## Electric-Field-Induced Electronic Instability in Amorphous Mo<sub>3</sub>Si Superconducting Films

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We report experimental evidence for the electric-field-induced electronic instability in the vortex state of superconducting amorphous Mo<sub>3</sub>Si films. At low magnetic fields, this instability results in an anticlockwise hysteresis of the voltage-current characteristics, as predicted by Larkin and Ovchinnikov, whereas at high magnetic fields the hysteresis is clockwise. To explain the unprecedented clockwise hysteresis, we propose that the inelastic quasiparticle scattering rate is higher after the electronic system is driven into the normal state by the electric field.

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Dynamic processes on a microscopic level play an essential role in most major phenomena in condensed matter physics. Of particular interest and importance in the case of superconductivity is the fact that these microscopic processes are strongly coupled to the macroscopic order parameter. This can be illustrated by the motion of the vortex lines (or, equivalently, of the order parameter) in type-II superconductors with a velocity  $v_L$  proportional to the electric field strength E. Because of the slow energy relaxation in (superconducting) metals, the distribution of the normal excitations at large  $v_L$  can deviate significantly from its equilibrium value. The shape of the quasiparticle distribution function in a superconductor strongly influences the superconducting order parameter which, in turn, determines the conductivity [1]. A remarkable possibility has been predicted by Larkin and Ovchinnikov (LO) [1]: The voltage-current (V-I) characteristics exhibit an abrupt hysteretic jump into a state with the normal-state value of the electrical resistivity. Unrelated to the Joule heating and depairing current  $i_0$ , the hysteretic jump in the LO theory occurs because at large vortex line velocities the number of quasiparticles diminishes inside the vortex cores and augments the outside. This causes a decrease of the vortex diameter and, correspondingly, of the viscosity coefficient with increasing vortex line velocity. At a critical velocity  $v_L^*$  the current as a function of the voltage attains a maximum, and, for the current-biased mode of experiment, there is a discontinuity in the V-I characteristics. This phenomenon is analogous to the "hot" electron transfer between two bands in semiconductors resulting in a negative differential conductivity dI/dV [2]. Although such a jump of the voltage has been observed experimentally in both low-temperature [3,4] and high-temperature [5] superconductors, no hysteresis has been reported to date.

In this Letter, we report novel phenomena associated with the electronic instability in the vortex state of amorphous- (a-) Mo<sub>3</sub>Si superconducting films: (i) The voltage exhibits a hysteretic behavior both as a function of the transport current I and of the applied magnetic field (*H*). (ii) Most importantly and interestingly, the hysteresis rotation is clockwise for the *V* vs *I* curves at high magnetic fields (or for the *V* vs *H* curves at low currents), in contrast to the anticlockwise rotation for the *V* vs *I* curves at low magnetic fields (or for the *V* vs *H* curves at high currents). To explain the clockwise direction of the hysteresis, we suggest that the inelastic electron scattering rate  $\tau_{\epsilon}^{-1}$  is hysteretic, being bigger on the descending branches of the *V*-*I* or *V*-*H* curves, after the excursion to the normal state.

This work consists of studies of three a-Mo<sub>3</sub>Si film samples; the results for them are all very similar. The zerofield transition temperature is 8.0 K (width < 20 mK). a-Mo<sub>3</sub>Si is an isotropic type-II superconductor, with the Ginzburg-Landau parameter ~60 [6] and  $j_0 \sim 10^7 \text{ A/cm}^2$ [7]. The films were rf sputtered on sapphire substrates as described in Ref. [6]; their dimensions are  $3 \text{ mm} \times$  $1 \text{ mm} \times 1700 \text{ Å}$ . Indium wires were attached to the samples for low resistance ( $<0.1 \Omega$ ) electrical contacts. The voltage was measured on the length  $l \approx 1$  mm along the largest sample dimension. The magnetic field H was always applied perpendicular to the substrate. The V-Imeasurements were done at T = 4.2 K. Direct contact with liquid helium and the high thermal conductivity of the sapphire substrate ensure highly effective Joule energy removal from the films. Therefore the electronic instability manifested by the V-I characteristics in this work is not of a thermal origin, as discussed in detail below.

Figure 1 shows the temperature dependence of the upper critical field  $H_{c2}$  as obtained from the low-current (Ohmic) resistivity measurements. For a fixed temperature,  $H_{c2}$  is determined as a field at which the resistivity R(H) (Fig. 1, inset) equals  $0.9R_n$  (with the normal-state resistance  $R_n = 4 \Omega$  for sample 1). The solid line represents a dirty-limit fit to the data [8] using the slope of the upper critical field at  $T = T_c(H = 0)\mu_0 dH_{c2}/dT = -2.5$  T/K. As exemplified in the inset of Fig. 1, the Ohmic response vanishes with decreasing field (at  $\approx 7.2$  T for T = 4.2 K).

The main panels in Fig. 2 illustrate the V-I curves, while the V vs H dependences are shown in the insets.

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FIG. 1. The plot of  $H_{c2}(T)$  for three Mo<sub>3</sub>Si samples. The solid line represents a dirty-limit fit [8]. Inset: The resistance R vs H for sample 1 at T = 4.2 K.

Let us start with the discussion of the V-I curves. At low fields (below  $\approx 4$  T, data not shown) the voltage is a completely reversible function of the applied current up to the highest values  $I_{\text{max}}$  used in our experiment (typically 40–50 mA). At higher magnetic fields, the curves are reversible until a certain value of the current, denoted by  $I_1$ , is exceeded. At this current a remarkable feature appears: an abrupt jump of the voltage to values close to  $R_n I$  (with  $R_n = 4 \Omega$  for sample 1) [Fig. 2(a),  $\mu_0 H = 5.27$  T]. Upon decreasing the current we observe an anticlockwise hysteresis, that is, the voltage drops at a current  $I_2 < I_1$ . Therefore V is an unambiguous function of I only if the current is in either one of the two reversible regions:  $0 < I < I_1$  and for  $I_2 < I < I_{\text{max}}$ .

Upon further increase of the magnetic field [Fig. 2(b),  $\mu_0 H = 6.23$  T], the hysteresis loop for *I* following the sequence  $0 \rightarrow 25$  mA  $\rightarrow 0$  [as shown in Fig. 2(b) by the solid squares] is more complicated than that for 5.27 T. The high-resistive part of the *V*-*I* curve is reversible only for I > 18.7 mA, while for  $I_1 < I < 18.7$  mA the voltage on the descending branch is lower than that for the ascending branch [see the inset in Fig. 2(b)]. Moreover, a "subloop" develops for  $I = 0 \rightarrow 16.2$  mA  $\rightarrow 0$ , as represented by the open circles in Fig. 2(b). We note that the value of 16.2 mA corresponds to a current immediately above  $I_1$ , and that the hysteresis width of the subloop is larger than that for the full loop.

The complicated behavior illustrated in Fig. 2(b) represents a crossover from an anticlockwise to clockwise hysteresis in the V-I curves with the increasing magnetic field. Indeed, at high fields [6.98 T, Fig. 2(c)] we arrive at an unprecedented, to the best of our knowledge, observation of a clockwise V-I loop, which is our main experimental result. For  $\mu_0 H = 6.98$  T the subloop  $0 \rightarrow 11.9$  mA  $\rightarrow 0$  (open circles) is narrower than the full loop  $0 \rightarrow 20$  mA  $\rightarrow 0$ (solid circles). With magnetic field increased above 7.5 T the (clockwise) hysteresis width vanishes; the discontinuity in V-I curves evolves into an inflection point which becomes less pronounced and shifts to I = 0 as the magnetic field approaches  $H_{c2}$ .

A similar hysteresis can be traced by keeping the current constant and recording V-H curves. For high (I =



FIG. 2. (a) V vs I for  $\mu_0 H = 5.27$  T.  $I_1$  ( $I_2$ ) is the current where the voltage jumps up (down). Inset: V vs H for I = 20 mA. (b) V vs I for  $\mu_0 H = 6.23$  T. The solid squares and arrows with solid ferrules are those for the loop  $0 \rightarrow 25$  mA  $\rightarrow 0$ . The open circles and arrows with open ferrules represent data for a subloop  $0 \rightarrow 16.2$  mA  $\rightarrow 0$ . Inset shows an extended view in the vicinity of the voltage jump. (c) V vs I for  $\mu_0 H = 6.98$  T. The solid squares and arrows with solid ferrules are those for the full loop. The open circles and arrows with open ferrules represent data for a subloop  $0 \rightarrow 11.9$  mA  $\rightarrow 0$ . Inset shows V vs H for I = 12 mA. The solid symbols are those for the full loop, while the open ones are for the subloop 8 T  $\rightarrow 6.875$  T  $\rightarrow 8$  T. For all main panels, the dashed lines represent a normal-state slope.

20 mA) current the hysteresis is located near  $\mu_0 H = 5.4$  T [Fig. 2(a), inset]; for low (I = 12 mA) current it occurs around  $\mu_0 H = 6.9$  T [Fig. 2(c), inset]. For high (low) currents the hysteresis direction is anticlockwise (clockwise) as shown in Figs. 2(a) and 2(c) insets.

Before further discussions of the physics, it is necessary to estimate the temperature increase  $\Delta T$  due to the Joule heating. We may equate the Joule power after the jump,  $I_1^2 R_n$ , to the surface heat transfer  $WS\Delta T$ , where W is the thermal conductance and  $S \approx 10^{-6} \text{ m}^2$  is the sample surface area. The surface thermal conductance is provided by the heat transfer to the surrounding liquid helium ( $\approx$  $10^4 \text{ W/m}^2 \text{ K}$  [9]) and to the sapphire substrate ( $\approx 2.5 \times$  $10^4 \text{ W/m}^2 \text{ K [9]}$ , so that  $W \approx 3.5 \times 10^4 \text{ W/m}^2 \text{ K}$ . For 5.27 T, the overheating after the voltage jump is  $\Delta T \approx$ 50 mK, while that for 6.98 T is  $\Delta T \approx 15$  mK. In both cases, the Stekly parameter [9]  $\beta = \Delta T / [T_c(H) - T] \approx$ 0.02. As  $\beta \ll 1$ , the heating effects are not important, and the generation of a "hot spot" [9] is proven not to occur. It is also worth noting that our main observation (that of the clockwise hysteresis) in any case cannot be explained by a thermal runaway. The fact that the instability in V-Icharacteristics (see below) is not triggered by the Joule heating is additionally supported by the observation that the energy dissipation VI where this instability occurs is not a constant. Rather, VI can vary by an order of magnitude by changing the magnetic field.

Having proven that the Joule heating is insignificant, we proceed to discuss the experimental results within the theory by Larkin and Ovchinnikov [1] which concerns an interesting nonequilibrium effect in the vortex dynamics. At large vortex line velocities, the electric field due to vortex motion results in a decreasing size of the vortex cores because quasiparticles accelerated by the electric field can reach energies above the superconducting energy gap and diffuse away from the vortex cores. Their effective pressure on the vortex walls drops and the vortex cores shrink. The vortex radius  $\xi$  and the viscous coefficient  $\eta$  are given by the following formula:  $\xi^2(V) = \xi^2(0)/[1 + (V/V^*)^2]$  and  $\eta(V) = \eta(0)\xi^2(V)/\xi^2(0)$ . The expression for the nonlinear V vs I dependence has the form

$$I = \frac{V}{R_n} \left[ \alpha \frac{1}{1 + (V/V^*)^2} + 1 \right].$$
 (1)

The characteristic voltage  $V^*$  is controlled by the inelastic electron scattering time  $\tau_{\epsilon}$ :

$$(V^*/l)^2 = \frac{1792\zeta(3)\beta_A k_B^2 (T_c - T)^3}{\pi^6 e^2 D T \tau_\epsilon},$$
 (2)

where  $\zeta(x)$  is the Riemann zeta function, *e* is the electron charge,  $k_B$  is the Bolzmann constant,  $\beta_A = 1.16$ ,  $D = (1/3)v_F l$  is the diffusion coefficient, with  $v_F$  the Fermi velocity, *l* the electron mean free path; and the coefficient  $\alpha$  given in Ref. [10].

The first term in the square brackets [Eq. (1)] describes the change in the conductivity due to renormalization of the viscosity  $\eta(E) \sim \sigma(E)$ , whereas the second term accounts for the suppression of the superconducting order parameter outside the vortex cores in the limit of a large electric field (see Ref. [11]). The *I-V* characteristic given by Eq. (1) acquires an N shape.

To illustrate this concept more quantitatively, we plot in Fig. 3 the V-I curves according to Eq. (1). The values of the parameters  $\alpha$  (14 for curve 1, 7 for curves 2) and  $V^*$  (7.3 and 9.6 mV for curves 1 and 2, respectively) were chosen to imitate the experimental data for  $\mu_0 H = 5.27$  and 6.98 T, respectively.

Let us consider first curve 1 in Fig. 3. One can see that I as a function of V has an N shape. In our experiment, the mode of operation is current biased. Therefore, as the current reaches the point where dI/dV would become negative for a V-biased mode, the system switches into the higher-resistive state as shown by the arrow. The electric field after the jump is high enough to destroy superconductivity, and the upper-voltage branch of the V-I curve is reversible [see Fig. 2(a) for the experimental data]. Similarly, with decreasing current, the voltage jumps down at a lower I than for jumping up, in agreement with the anticlockwise hysteretic data at 5.27 T [Fig. 2(a)]. However, if the magnetic field increases further, the coefficient  $\alpha$  decreases [1,10], so that the voltage jump becomes less pronounced, in agreement with the data for 6.98 T [Fig. 2(c)]. The LO theory predicts an unambiguous relationship between V and I for large enough magnetic fields (curve 2, Fig. 3), where we observe the clockwise hysteresis.

To account for the clockwise hysteresis, we consider the following possibility. After the jump to the higher-resistive state the order parameter vanishes, and the electron-electron (inelastic) scattering rate increases. Therefore, at decreasing current from the normal state, the characteristic electric field  $E^* \sim \tau_{\epsilon}^{-1/2}$  is larger than on the ascending branch from the superconducting state. By taking the hysteretic scattering rate into account, we obtain curves 2 and 3 with the same value of  $\alpha$  and with different  $V^*$  values (9.6 mV for curve 2 and 9.8 mV for curve 3) to represent a larger scattering rate for curve The widest clockwise loop in the high-H limit can 3. be achieved if the system is driven completely into the normal state. Otherwise (i.e., in case of a subloop), the increase in  $V^*$  for the descending branch is smaller than for the full loop, and the clockwise subloop is narrower than the full one [Fig. 2(c)]. The maximum change in the energy relaxation time (for the full clockwise loop) is estimated to be  $|\Delta \tau_{\epsilon}/\tau_{\epsilon}| \sim 2|\Delta V^*/V^*| \sim$  $2(0.2 \text{ mV}/9.6 \text{ mV}) \approx 0.04 [12].$ 



FIG. 3. The V-I curves as given by Eq. (1). See the text for values of parameters. Inset:  $I_c$  vs H.

Similarly, the V-H curves at a fixed current I can be understood in terms of the analysis given in Fig. 3. By inspecting Fig. 3, one can easily find out what V-H curves at fixed current I one can expect. For large (small) driving currents, the region where voltages change significantly occurs at low (high) magnetic fields. The (anti)clockwise hysteresis in the V-I curves results in the same direction of the hysteresis in the V vs H dependences, in agreement with the experimental results [see Figs. 2(a) and 2(c), insets].

We would like to make several remarks concerning the applicability of the LO theory. The numerical coefficients in Eqs. (1) and (2) and in Ref. [10] should be considered as an order-of-magnitude estimate [1] because of the oversimplifications made in the theoretical model (see Ref. [11], for instance). By using the theoretical expression for  $\alpha$  in the low-*H* limit [10], we obtain  $\alpha \approx 15$ for  $\mu_0 H = 5.27$  T, consistent with the parameter  $\alpha = 14$ used for curve 1 in Fig. 3. Similarly, we obtain  $\alpha \approx 4$  for  $\mu_0 H = 6.98$  T if the theoretical expression in Ref. [10] for  $\alpha$  in the high-*H* limit is employed. This value is in reasonable agreement with the parameter  $\alpha = 7$  used in curve 2 of Fig. 3, considering the oversimplifications in the theory.

From Eq. (2), using the measured value of  $v_L^* = E^*/\mu_0 H \approx 1$  m/s together with the diffusion coefficient determined from the slope of the upper critical field [8]  $D = (4k_B/\pi e) (-dT_{c2}/\mu_0 dH)|_{H=0} \approx 4 \times 10^{-5}$  m<sup>2</sup>/s, we can estimate the inelastic electron scattering time  $\tau_{\epsilon} \sim 10^{-7}$  s (see Ref. [13] for comparison with other previous work [3–5]).

Another noteworthy point is that the LO theory [1] does not take into account pinning, which is always present in real systems. Shown in the inset of Fig. 3 is the plot of the critical current  $I_c$  needed to depin the vortices (with a criterion of 50 nV corresponding to an electric field 50  $\mu$ V/m). (The value of 5 mA corresponds to the current density of about  $3 \times 10^3$  A/cm<sup>2</sup>  $\ll j_0$ .) In the first order approximation, a nonzero critical current  $I_c$  would shift the whole picture of dissipation to higher currents, thereby providing a better agreement between the data in Fig. 2 and the theoretical curves in Fig. 3.

To conclude, we have attributed the observation of novel current-voltage characteristics in the vortex state of a-Mo<sub>3</sub>Si films to nonequilibrium effects associated with the electric-field-induced electronic instability. At low magnetic fields this instability results in an anticlockwise hysteresis in the *V*-*I* and *V*-*H* curves as predicted by the LO theory. At high magnetic fields, we have observed a novel phenomenon in the nonequilibrium physics of superconductors: a clockwise hysteresis which may be attributed to the increasing of the inelastic electron scattering rate as the superconductivity is suppressed by the presence of large electric field.

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- [10] In the low-field limit the coefficient  $\alpha$  is  $\alpha = 4.04H_{c2}/(1 T/T_c)^{1/2}H$ . For magnetic fields close to  $H_{c2}$  (Ref. [1(a)])  $\sigma/\sigma_n = 1 + (\pi^2|\psi|^2/32T)[T_c(E) T_c]^{-1}$ , with the order parameter  $|\psi|^2 = [2\pi^3k_B \times TeD/7\zeta(3)\beta_A]$  ( $-[dT_{c2}/d(\mu_0H)|_H]^{-1})/[1 + (V/V^*)^2]$ . This leads to  $\alpha = [\pi^5 ek_BD/112\zeta(3)\beta_A]\mu_0(-dT_{c2}/dH|_H)^{-1}$ .
- [11] In fact, the suppression of the order parameter outside the vortex cores due to the quasiparticles coming from the cores should be taken into account by revising Eq. (1) into  $I = (V/R_n) (\alpha/[1 + (V/V^*)^2] + f(E))$ , where the function f(0) = 0 and asymptotically approaches 1 at large *E*. Data for 5.27 T imply that  $f(E) \approx 1$  at  $E \approx 85 \text{ V/m}$  ( $V \approx 85 \text{ mV}$ ). The functional form may be related to the Fermi-Dirac function and needs further theoretical investigations.
- [12] The electron-electron scattering rate is proportional to the normal electron density n(H). The relative change in n when the system is driven into the normal state is  $\Delta n(H)/n \sim 1 - H/H_{c2} \approx 0.15$  for 6.98 T. The fact that  $\Delta n(H)/n > \Delta \tau_{\epsilon}/\tau_{\epsilon}$  may be the result of the following: At a given location, the relaxation of the quasiparticle distribution and of the order parameter between the passing high density vortices will be incomplete (see Ref. [4]). In contrast to the limit of large vortex separation, the effective relaxation time can be prolonged significantly. Consequently, at large H the values of both  $V^*$  and  $v_L^*$  are reduced according to Eq. (2)  $(V^* \sim \tau_{\epsilon}^{-1/2})$ , and so is the ratio  $\Delta \tau_{\epsilon}/\tau_{\epsilon}$ . With H approaching  $H_{c2}$ ,  $\Delta n(H)/n$  vanishes, and so does the hysteresis in  $\tau_{\epsilon}$ . (The hysteresis width drops below our experimental resolution between 7.5 and 8 T.)
- [13] For all the samples of Refs. [3,4] (In and Al films),  $D > 3 \times 10^{-4} \text{ m}^2/\text{s}$ ,  $\tau_{\epsilon} < 3 \times 10^{-8} \text{ s}$ ,  $v_L = 600 10^4 \text{ m/s}$ . For YBCO [5],  $D \approx 1.7 \times 10^{-4} \text{ m}^2/\text{s}$ ,  $\tau_{\epsilon} \approx 2 \times 10^{-8} \text{ s}$ ,  $v_L \approx 80 \text{ m/s}$  (at T = 50 K).