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Frequency dispersion of nonlinear response in films of cuprate superconductors

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The effects of microwave radiation on transport properties of atomically thin $La_{2-x}Sr_xCuO_4$ films were studied in the 0.1-20 GHz frequency range. Resistance changes induced by microwaves were investigated at different temperatures (8−15 K) near the superconducting transition. A strong decrease of the nonlinear response is observed within a few GHz of a cutoff frequency $\nu_{cut} \approx 2$ GHz. The expected frequency dependence vastly underestimates the sharpness of this drop. Numerical simulations that assume ac response to follow the dc *V-I* characteristics of the films reproduce well the low frequency behavior, but fail above ν_{cut}. Thus, high-frequency radiation is much less effective in inducing vortex-antivortex dissociation in the oscillating superconducting condensate.

Transport properties of thin superconducting films have attracted much interest due to a fascinating physical phenomenon, the Berezinskii-Kosterlitz-Thouless (BKT) transition, predicted to occur at the critical temperature T_{BKT} lower than the temperature of the superconducting transition in bulk samples. [1-3] The BKT phase transition originates from long-range (logarithmic) interactions between vortex excitations in a two-dimensional (2D) superconducting condensate. Below T_{BKT}, the dominant thermal excitations are vortex-antivortex (V-AV) pairs. Subcritical (i.e. superconducting) current does not move pairs and hence does not dissipate energy. However, a stronger current can break V-AV pairs, generate free vortices, set them in motion via the Lorentz force, and thus make the transport dissipative. [4,5] The induced dissociation of V-AV pairs depends on the current strength and thus results in an extraordinary violation of the Ohm's law. With this motivation, the strong nonlinear response to the electric current in thin superconducting films was investigated extensively. [6-19] Despite significant progress, some key questions are still being debated. In particular, a recent theoretical study [17] indicates that weakening of

V-AV pairs near the edges of a superconducting film may provide a substantial, and perhaps even dominant, contribution to the nonlinear *dc* response. Note that the majority of studies of the nonlinear transport near the superconducting transition in thin films have in fact been done in the *dc* domain.

This Letter presents experimental investigations of nonlinear transport properties of atomically thin $La_{2-x}Sr_xCuO_4$ films over a broad frequency range, $0 \leq \nu \leq 20$ GHz. The experiments indicate a dramatic decrease of the nonlinear response at high drive frequencies, suggesting significantly reduced effectiveness of high-frequency radiation to dissociate V-AV pairs in the oscillating superconducting condensate.

The experiments were performed on thin $La_{2-x}Sr_xCuO_4$ (LSCO) films synthesized by Atomic-Layer-by-Layer Molecular Beam Epitaxy (ALL-MBE). [19-30] This is the premier technique for growth of atomically perfect, single-crystal high temperature superconductor (HTS) films. By virtue of "digital" synthesis, ALL-MBE also allows fabrication of precise multilayer heterostructures that contain ultrathin layers, down to thickness of one unit cell (1 UC). On the extreme level of control, delta-doping in a single CuO_2 layer has been demonstrated. [21] This unique synthetic capability has been essential for the present work.

The profile of the Sr doping level was modulated as follows. The film A is a heterostructure with three distinct layers, each of which is exactly 5 UC thick. (In LSCO, the unit-cell height is 1.32 nm.) The top and the bottom layers were made of $La_{1.59}Sr_{0.41}CuO_4$ (M) which is strongly overdoped and metallic but not superconducting. The inner slab is a 5 UC thick layer of $La_{1.79}Sr_{0.28}CuO_4$, which is a superconductor (S) with $T_c \approx 7$ K. The film B has a similar M-S-M structure but the superconducting part is much thinner, 1.5 UC thick, and made of $La_{1.80}Sr_{0.20}CuO_4$. While the Sr doping level in the central S layer of sample B is lower than in the sample A, the actual hole densities are similar. In the heterostructure B, the hole density in the central layer is larger than the Sr doping level because of significant charge accumulation in S and depletion in M near the two M-S interfaces, so here $T_c \approx 6$ K. In the bottom insert in Fig. 1, the temperature dependence of resistivity is shown for both samples.

The films were lithographically patterned into the shape of Hall-bar devices with the width $W = 200$ μm and the distance between the voltage contacts $L = 800$ μm. The resistance was measured using the four-point probe method. A direct electric current (*dc* bias, I) was applied through a pair of current contacts, and the longitudinal *dc* voltage V was measured between the potential contacts. The microwave (MW) and radio frequency (RF) radiation was guided by a rigid coaxial line and applied to the samples electrically connected to the end of the coax as shown in the upper insert in Fig. 1. A 50 Ω microwave resistor terminates the end of the coax reducing significantly the reflected-back RF/MW power and providing for the broadband matching of the RF/MW circuit. A capacitor $C = 47$ pF and low pass filters (LPF, F < 100 MHz) separate *dc* and RF/MW circuits as shown in the insert.

The measurements were done in two regimes. In the first regime, a dependence of the voltage V on the bias *dc* current I was measured while a continuous RF/MW radiation was applied. In the second regime, the measurements were done using a lock-in amplifier locked with power modulation. The RF/MW power was modulated at the frequency $\nu = 15$ Hz and the variation of the voltage ΔV was measured as a function of the *dc* bias I. The samples and a calibrated thermometer were mounted on a cold copper finger in vacuum. The measurements were carried out at temperatures in the range 4.5 K ≤ T ≤ 20 K.

Figure 1 shows the dependence of the voltage V on the current I taken at different powers of the RF radiation as labeled. The RF radiation increases the resistance of the sample B as shown by thin solid lines. The thick solid line presents the $V-I$ dependence with no radiation applied. The open circles show the results of a numerical simulation of the response to RF radiation. The simulation assumes that the electromagnetic response of the sample follows the nonlinear $V-I$ dependence obtained in the *dc* domain. Since the response is nonlinear, the time averaged voltage $< V(t) >$ and current $< I(t) >$ that yield the open circles in Fig. 1 differ from the original values V_0 and I_0. Although the RF voltage $U_{RF}(t)$ at the end of the coaxial line is known, the value of the RF current $I_{RF}(t)$ flowing through the sample is not. This current is determined by the coupling between the sample and the RF circuit, which, in the RF setup employed here, depends on the high-frequency contact resistance R_c between the RF current leads and the film, which is not known accurately. Thus, we used R_c as the single fitting parameter in our simulations, and this was sufficient to achieve a very good agreement between the simulation and the experiments. At small currents, both the experiment and the simulations display a linear relation between the current I and voltage V – Ohm's Law.

Figure 2 presents the dependence of the sample resistance on microwave power taken at different temperatures as labeled. The open dots are the slopes of the $V-I$ dependences taken at small currents at which the linear (Ohmic) relation between the voltage V and the current I is observed. The variation of the resistance with the applied MW power depends considerably on the temperature. Close to the superconducting state, where the resistivity is small (at T = 6-7 K), the microwave-induced resistance variations are strong, whereas near the normal state (T = 8-9 K) the variations are weak. An additional feature of the evolution with the temperature is a change in the shape of power dependence. In particular, at T = 6 K this dependence looks more linear than the one seen in the data taken at T = 6.5 K.

The shape of power dependence shows more dramatic changes in the response at different frequencies. Figure 3 shows the variations of resistance from RF/MW radiation at different frequencies (0.17 ≤ ν ≤ 5 GHz) as labeled, taken at T = 7 K. At low frequency (ν = 0.17 and ν = 0.37 GHz), the response is in a reasonable agreement with the numerical simulations based on the nonlinear *dc* V-I curve. At higher frequencies, the nonlinear response deviates significantly from the low-frequency behavior, indicating that

at higher frequencies the response does not follow the *dc V-I* curve. At frequencies above 3 GHz, the nonlinear response is significantly weaker then the one observed at low frequencies at the same RF/MW power. However, the strength of the nonlinear response is restored at a higher applied power, as can be seen from the power dependencies taken at ν = 1.5 GHz and ν = 3 GHz.

Further investigations were done in the second regime (power modulation, lock-in amplifier) that facilitates measurements of small MW-induced deviations from the *dc V-I* curve. Figure 4 shows the dependence of the RF/MW-induced variations of the *dc* voltage ΔV on the *dc* bias current I taken at different frequencies and power levels as labeled. At a small *dc* bias, the response is proportional to the current, indicating no observable MW/RF rectification in the device. This is further supported by observations of complete anti-symmetry of the nonlinear response with respect to the *dc* bias (not shown). The left panel shows the effect of the low-frequency RF radiation (ν = 0.575 GHz) on the sample resistance. All three curves are in very good agreement with the ones obtained by numerical simulations for the same applied RF power levels as labeled. With the only open parameter in the model fixed at a particular value, R_c = 62 Ω, the simulations replicate all the details of the experiments including the shift of the observed maximum with increased RF power. The right panel shows the effect of the high frequency (ν = 3.4 GHz) radiation on the resistance. One can see that at high frequencies it takes a significantly higher MW power to achieve a comparable change in the resistance. Furthermore, the numerical simulation fails to fit the experiment at high *dc* biases even if one tries to adjust the magnitude of MW current, clearly indicating that the high-frequency response does not follow the *dc* V-I curve.

Figure 5 displays the behavior of the linear part of the $\Delta V(I)$ dependence on the RF/MW power at different frequencies as labeled. At small power, the slope of $\Delta V(I)$ is proportional to the power, as indeed expected (see below). At higher powers, the dependence is weaker. At higher frequencies, the transition to weak power dependence occurs at a higher power. At the highest power levels, all dependencies converge. Similar behavior is seen in Fig. 3.

An analysis indicates several distinct features of the *dc* nonlinear response presented in Fig. 1: finite Ohmic resistance at small currents, a cubic dependence at intermediate currents, and a $V \propto I^{\alpha}$ response at high currents with the exponent $\alpha(T)$ = 1−8, decreasing as the temperature increases. The dependence is presented below:

$$V = R_0 I + [\, \gamma I^3 \rightarrow \beta I^{\alpha} \; (I \rightarrow \infty) \,] \qquad (1)$$

where R_0 is the Ohmic resistance, the coefficient γ captures the dominant nonlinear mechanism at small currents, and β is a constant. The high-current behavior is in agreement with the one expected within the BKT scenario. [4,5] At low currents, the nonlinearity is well described by the cubic term in Eq. (1) which captures all significant features of the low frequency response presented in Figs. 4 and 5. Indeed at a small RF current $I_\nu(t)$, the voltage variations $\Delta V(I + I_\nu) = \langle V\,(I + I_\nu(t)) \rangle - V(I) = 3\, I \langle I_\nu(t)^2 \rangle$ are

proportional to the *dc* bias current *I*, which agrees with Fig. 4, and proportional to the RF power $P \sim I_\nu^2$, which agrees with Fig 5. The brackets < > indicate the time average. The decrease of the nonlinear response shown in Fig. 4 at high *dc* bias, and the transition to a weaker power dependence presented in Fig. 5, are the results of the term βI^α in Eq. (1) for $\alpha < 2$. However, the high-frequency behavior cannot be completely described by Eq. (1). Although the high-frequency response at $\nu = 3.4$ GHz is proportional to the *dc* bias at $I < 25$ μA, the transition to the high *dc* bias regime occurs at considerably higher current *I*, which is inconsistent with the *dc V-I* dependence.

Fig. 6 presents the frequency dependence of the effect of RF/MW radiation on the resistance, taken at different temperatures as labeled. The dispersions were measured in the regime of small currents and powers, at which the response is proportional to the *dc* bias current *I* and to the RF/MW power *P*. At low temperatures, $9 \leq T \leq 11$ K, the response to the RF/MW radiation decreased by 3-4 orders of magnitude, as the frequency was increased. At a high temperature, $T = 18$ K, the frequency dependence of the response gets much weaker. This weak frequency dependence is compatible with a heating mechanism of the nonlinearity proposed to occur at higher temperatures [Ref]. At low temperatures, the observation of a strong frequency dispersion of the nonlinear response points towards a mechanism related to superconductivity in the sample.

Considerable frequency dispersion may be expected to occur within the BKT picture. The V-AV pair dissociation depends on the strength of the driving electric current. Due to inertial properties of the condensate, the superconducting current is inversely proportional to the frequency ν of the electric field (inductive response). Since at a small *ac* current I_ν the nonlinear response is proportional to I_ν^2, one may expect $\Delta V \propto 1/\nu^2$. Numerical simulations indicate a considerable effect of the contact resistance on the applied RF current. The presence of the contact resistance in series with the inductive sample diminishes the $1/\nu^2$ frequency dependence. At frequencies above the threshold $\nu_{cut} \approx$ 2-3 GHz, the frequency dispersion shown in Fig. 6 is considerably stronger (proportional to $1/\nu^7$) than the inverse-square law. The fact that the reduction of nonlinear response is large indicates that the dominant mechanism of nonlinearity is involved. Within the BKT theory, the main contribution should come from current-induced dissociation of V-AV pairs. Thus, the observed behavior indicates that high-frequency oscillations in the superconducting condensate are significantly less effective in dissociating V-AV pairs.

In support of this suggestion, we note that, in contrast to the *dc* response, an oscillating super-current induces an oscillating motion of the vortex (antivortex) inside a V-AV pair. If in the course of oscillation the distance between the vortex and antivortex ($\propto 1/\nu$ for a viscous vortex motion) within a V-AV pair does not exceed a critical distance l_c, the pair can survive and will not be driven by the *dc* bias current. Thus, the high-frequency radiation may not affect the superconducting response. Another important observation - the convergence of the nonlinear response at high power levels shown in Fig. 5 and Fig. 6 - can also be

understood within the same scenario. At high power, the amplitude of the oscillating V-AV separation exceeds the critical distance l_c and the pairs are "broken" for at least some time during the RF/MW period, making the high-frequency nonlinear response similar to the one observed at low frequencies. Thus, the proposed mechanism provides a qualitative description of the observed behavior. The critical length l_c can be related to the screening length of the V-AV interaction l_{scr} [8] and/or to the tunneling time through the pair potential barrier modified by the radiation [32].

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FIGURE CAPTIONS

Figure 1: (Color online). Dependence of the voltage V on current I at different RF power applied to a 50 Ohm resistor at frequency ν = 370 MHz and temperature T = 7 K. The thick solid line represents the dc response with no RF power applied. The open circles are the results of numerical simulations of the effect of RF radiation on $V-I$ curve computed at the same RF power, for the contact resistance R_c = 67 Ω in series with the sample. The upper-left inset: a schematic of a dc circuit isolated from the RF circuit by a capacitor C = 47 pF and three low-pass filters (LPF, F < 100 MHz). The lower-right inset: resistance of the samples A and B as a function of temperature.

Figure 2: Resistance of the sample B as a function of applied MW (ν = 1.5 GHz) power at different temperatures as labeled.

Figure 3: (Color online). Resistance of sample B at T = 7 K as a function of the applied RF/MW power at various frequencies (open circles with dotted lines), showing strikingly different behavior for frequencies above 1.0 GHz. The thick black line is the dependence simulated assuming that R_c = 67 Ω and that RF/MW excitations follow the dc V-I characteristic.

Figure 4: (Color online). The change of voltage $\Delta V = V(P) - V(0)$ due to the applied RF/MW power P as a function of the dc bias current I at different microwave powers as labeled. The solid lines are experimental data; the open circles are results of numerical simulations. A comparison of the left and right panels shows the agreement between experiment and simulation at low frequencies, but serious disparity at high frequencies. All the experimental data were taken from sample A at T = 11 K; all the simulations used the fitting parameter R_c = 63 ± 2 Ω.

Figure 5: (Color online) The dependence of the linear part of response, $\Delta V(I) \sim I$, in sample A on the applied RF/MW power, observed at small *dc* biases in Fig. 4, at T = 11 K and at different frequencies as labeled. The solid line is the dependence $\Delta V \sim P$ expected for small power P. The open circles are the results of numerical simulations, assuming R_c = 62 Ω.

Figure 6: The frequency dependence of relative variations in resistance of sample A normalized by the applied RF/MW power at different temperatures as labeled. The dashed line approximates the decrease of the nonlinear response for 3 GHz ≤ ν ≤ 6 GHz. The solid line is the expected frequency dependence taking into account the inertial properties of superconducting condensate.