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### Voltage Responses to Optical Pulses of Unbiased Normal and Superconducting Samples

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# Voltage responses to optical pulses of unbiased normal and superconducting samples

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The direct transformation of the energy of an incident high-energy photon into a measurable potential difference within an absorbing metal is investigated. Experimental evidence is presented that the effect arises from the inherent energy dependence of the electronic density of states, rather than from a simple temperature excursion. The similarities between the results on Al and YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> samples indicate that the effect is universal in nature. We assert it may be used as the basis of a fast, energy resolving, individual photon detector for the ultraviolet radiation and x-rays.

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In Ref. 1 it was suggested on theoretical grounds that the nonequilibrium response of superconductors has the potential to be used as the basis of a high energy resolution detector. The transient response of superconductors to light pulses was earlier studied by many authors (see the recent review<sup>2</sup>), in order to measure the inherent time scales of the relaxation processes. Most of this work was performed in the presence of a bias current. Thus, the main voltage signal arose from the impedance change associated with the temperature shift. However, both Refs. 3 and 4 mentioned observing a nonzero signal at zero bias current. In addition, there is evidence of the same effect in studies of the thermopower of YBCO (YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>).<sup>5,6</sup>

When a high energy quantum is absorbed in the metal, a nonequilibrium state is created; the electronic distribution function  $f(\epsilon)$  is at first not describable by the Fermi-Dirac formula  $f(\epsilon) = \{1 + \exp[(\epsilon - \mu)/T]\}^{-1}$ , because a local temperature is not yet well defined. Rather the electron distribution has a spike at high energy which evolves in time downward in energy space, increasing the number of excess electronic excitations. Between points in the equilibrium and nonequilibrium regions a potential difference exists and can be measured experimentally. The value is the difference between the electrochemical potential  $\mu$  in the equilibrium and nonequilibrium states. One can prove that  $\mu$  in both cases could be determined by the condition  $f(\epsilon = \mu) = 1/2$ . The difference in electrochemical potentials can be measured without applying a bias current. In the superconducting state, the gauge invariant component of the electrochemical potential shift may also be registered at a single point in the disturbed volume, as a potential difference between normal and

superconducting state leads.<sup>1</sup> Herein we report demonstrations of these detector concepts.

The YBCO samples were all 0.5- $\mu\text{m}$ -thick, polycrystalline,  $c$ -oriented, and entirely unpatterned. Grown on MgO substrates, the typical  $T_c$ 's were  $\sim 90$  K. One set of experiments was made using a Cr:LiSrAlF<sub>6</sub> pulsed laser with a cavity  $Q$  switch that produces 40 ns duration pulses with a few Hz repetition rate. The fundamental frequency in the near-IR could be doubled and quadrupled using the nonlinear crystals LBO (LiB<sub>3</sub>O<sub>5</sub>) and BBO ( $\beta$ -Ba<sub>2</sub>BO<sub>4</sub>), respectively. This capability was utilized to look for evidence of the non-thermal nature of the measured voltage. If the response was purely thermal, then only the total energy delivered to the sample should influence the magnitude of the voltage measured. Our nonequilibrium model<sup>1</sup> suggests, however, that the higher the energies of the individual incident photon, the higher the value of  $\mu$ -potential shift. Figure 1 compares signals produced by the different harmonics of the laser, where the photon fluences were adjusted to deliver the same total energy in the range from 0.1 to a few mJ/cm<sup>2</sup>. The experimental reality is clearly that the response increases with the photon frequency. In this set of measurements the samples were biased at room temperature.

Figure 2 elucidates the lower limit of the time-resolution capabilities of the YBCO-based prototype detector. An acousto-optic cell was used to produce a series of subnanosecond pulses. The voltage observed from the YBCO has essentially the same wave form as the light itself, indicating that the inherent relaxation rate of YBCO is no slower than these pulses. We conclude, thus, that a potential counting rate capability of better than 1 quantum/10 ns has been demonstrated. The contemporary understanding of the nonequilibrium dynamics in YBCO indicates<sup>2</sup> that the relaxation time  $\tau_\epsilon$  of charge carriers may be much shorter:  $\tau_\epsilon \sim 1 - 10$  ps. Thus, much higher counting rates should be pos-

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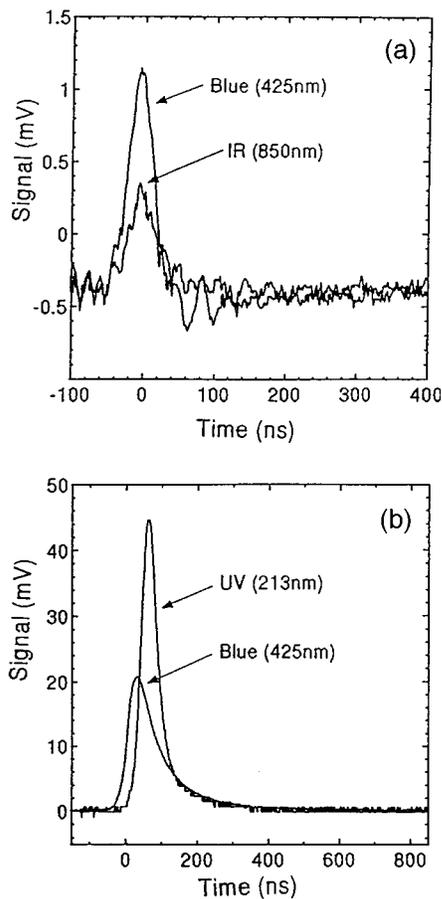


FIG. 1. Signals for different light wavelengths at the same amount of deposited energy: infrared vs blue for energy density  $\sim 0.1 \text{ mJ/cm}^2$  (a) and, in a different sample, blue vs ultraviolet for energy density  $\sim 10 \text{ mJ/cm}^2$  (b).

sible if the duration of  $\mu$ -potential response is determined by  $\tau_\epsilon$ .

Our results on Al films confirm this expectation. 40-nm-thick Al films were deposited on (001) semi-insulating (resistivity  $> 50 \Omega \text{ cm}$ ) Si wafers. An  $\text{Ar}^+$  ion laser ( $\lambda = 0.488 \mu\text{m}$  line) pulses were formed by passing the continuous wave beam through an acousto-optic modulator. A  $3\text{-}\mu\text{m}$ -wide stripe etched into a serpentine shape formed a sensor, covering  $50 \times 75 \mu\text{m}^2$  area, that was then mounted in a  $^3\text{He}$  cryostat with optical windows. The laser was focused on the sample surface to produce a typical spot size of  $20 \mu\text{m}$  in diameter. With the help of a microscope, the position of the laser spot with respect to the Al sensor could be easily determined. Special microaxial cables were used to ensure a  $50\Omega$  transmission line impedance and restrict heat flow to the sample. The stainless steel outer conductor and Nb/Ti inner conductors were connected to opposite ends of the sensor. This produced a dc thermoelectric current in the sensor. The laser pulse width was  $\sim 10 \text{ ns}$ , and the peak power delivered to the sample was  $0.5 \text{ mW}$ . The results when one end of the sensor strip is illuminated are summarized in Fig. 3. Notice that in this case the fast peak has a duration of  $100 \text{ ns}$ , far longer than the  $10 \text{ ns}$  laser pulse which produces it. This contrasts strongly with the case in YBCO where the light pulse and response had the same duration within experimental error. We interpret this distinction as directly reflecting the more than 3 orders of magnitude slower inherent relax-

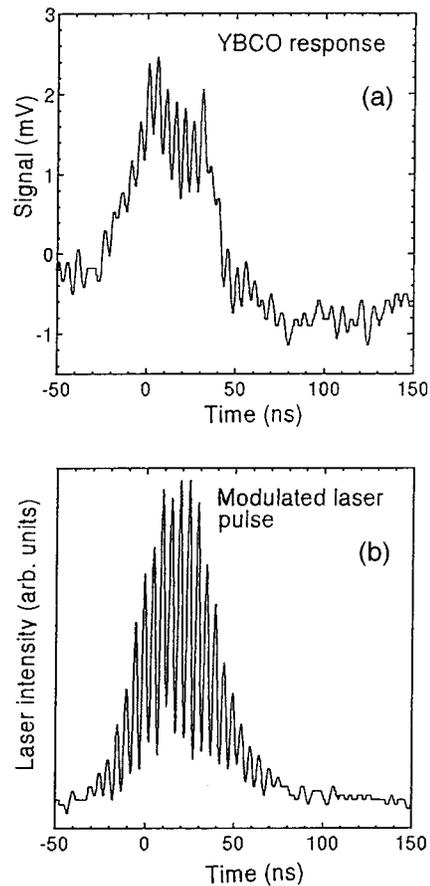


FIG. 2. Temporal shape of the laser pulse as measured by the YBCO-based prototype detector (a) and by a silicon photodiode (b).

ation times in Al than in YBCO. The fast peak reverses in sign as it should, when the opposite end of the serpentine is illuminated and goes to zero in the middle. Moreover, the fast components of the response have the same order of magnitude in the superconducting and in the normal states (the ambient temperatures in both cases were of the order of  $T_c$ ). The slow signal in Fig. 3 has a sign that is independent of the position of the spot illuminated. It is caused by the sample undergoing a superconducting to normal phase transition. While in the normal state, the sample's resistance becomes nonzero and the cable-caused bias current then produces a

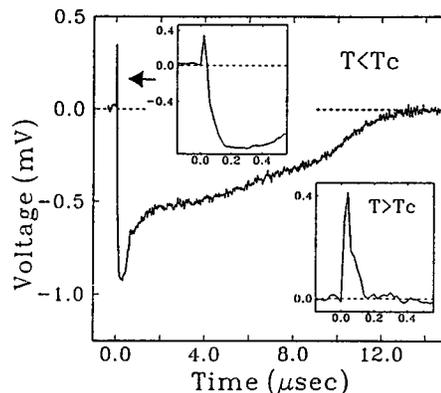


FIG. 3. Voltage signals from Al samples. Temperatures below and above  $T_c$  ( $= 1.21 \text{ K}$ ) are  $0.38$  and  $2.42 \text{ K}$ , respectively.

Joule heating voltage drop. The duration of this pulse is dominated by this heating and phonon trapping, which slows the sensor's cooling and return to the superconducting state.

The experiments on YBCO did not have this bias current which, in the Al case, provides clear evidence of a local conversion of the superconductor into the normal state and back. The YBCO pulsed laser radiation measurements were performed at temperatures ranging from 10 K below  $T_c$  to room temperature. Typical power densities in the pulses were  $\sim 10^4$  W/cm<sup>2</sup>. While this laser intensity was much larger than in the Al experiments, the transition temperature (and appropriate pairing energies) are also  $\sim 2$  orders of magnitude larger in YBCO than in Al. Lower intensity radiation must be used to distinguish between superconducting and normal samples.<sup>7</sup>

One can make some estimates related to the sensitivity  $S$  [V/(J/vol)] of the proposed detector scheme for the UV radiation case. For specificity, consider  $A = 10 \times 10 \mu\text{m}^2$  pixel and utilize the responsivity of the sample in Fig. 1(b) to 213 nm radiation. Such a pixel produces a  $U = 50$  mV pulse in response to a 10 nJ energy absorbed in a 500-nm-thick film. The film need only be  $d = 10$ -nm-thick for effective  $E_0 = 6$  eV photon absorption. Thus, a  $U = SE_0/dA \sim 0.25$  nV signal would be expected in response to  $E_0 = 6$  eV =  $10^{-18}$  J energy. However, this strongly underestimates the response amplitude. The  $\tau_0 = 40$  ns long pulse in Fig. 1(b) represents an effective steady state for the YBCO. Only during the initial time interval is the amplitude rising. Once this value is achieved, the rest of the energy delivered goes only to maintain this value against the energy relaxation (see Fig. 2). The rate of energy relaxation is temperature dependent and much higher at higher temperatures. In YBCO it scales linearly in  $T$  and at room temperatures, to which case our data refer, the relaxation time  $\tau_e$  of charge carriers should be not longer than 1 ps. So in the case  $\tau_0 \gg \tau_e$ , the sensitivity actually is a factor of  $\tau_0/\tau_e$  larger and the predicted 6 eV response of 0.25 nV should be enhanced at least by a factor of  $4 \times 10^4$ . Our experimental results using pulses with  $\tau_0 < \tau_e$  with Al confirm this prediction, once the impact of much smaller  $\omega_0/\epsilon_F$  on the mechanism of creation of the nonequilibrium potential and the partial transparency of the Al film used are recognized.

A virtue of our scheme of single photon detection is that at lower temperatures, the lifetime of produced signal will be substantially longer. For YBCO,  $\tau_e \sim T^{-1}$ . Photon absorption is instantaneous on this time scale. Thus, one can expect that a single 6 eV UV photon will produce at least 10  $\mu\text{V}$  voltage signal in a single  $10 \times 10 \mu\text{m}^2$  YBCO pixel, which will relax in tens of ps at 4 K.<sup>2</sup>

This signal amplitude must be compared to the Johnson voltage noise. At 4 K the magnitude of such noise expected in an  $R = 1 \Omega$  sample over a 100 ps time scale is 1  $\mu\text{V}$ . So one expects a signal-to-noise ratio of 10. If the pixel is in the superconducting state, the sample resistance will be determined entirely by the contact resistance which can be made as small as  $10^{-8} \Omega \text{cm}^2$  for YBCO to gold joints, that is,  $R = 0.01 \Omega$ , with a signal-to-noise ratio even one order of magnitude better. Whether the resistance can be further decreased, and higher accuracy achieved by using a second superconductor to carry the voltage signals out from the small pixels to larger contact pads is under investigation. We conclude that there are positive prospects for this new class of fast detectors based on perturbations in the electrochemical potential.

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