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Far-infrared photoresponse of quasi-two-dimensional granular NbN/BN films

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The photoresistive response to pulsed far-infrared radiation has been measured for a thin quasi-two-dimensional film of granular NbN/BN near the resistive onset of the film. The photoresponse dependence on temperature and light intensity is observed to be a strong function of whether the energy of the far-infrared photons is less or greater than the superconducting gap energy of bulk NbN. The results are consistent with a mechanism of direct coupling to intergrain Josephson currents that dominates for below-gap light and depairing processes (vortex-antivortex or Cooper pairs) for above-gap light.

Thin films of granular NbN/BN consist of small grains (~ 100 Å in diameter) of superconducting NbN dispersed in an insulating BN medium.[1] The transition to the zero-resistance state of these films can be described in terms of a Kosterlitz-Thouless quasi-two-dimensional (2D) model,[1-4] where the 2D transition to zero resistance at the temperature T_c is dominated by vortex motion. The individual NbN grains of the film have a superconducting transition temperature T_{c0} ($T_c < T_{c0}$).

Under current bias near T_c these films have been shown[5] to exhibit a photoresponse (PR) to visible light which is peaked near the 2D transition temperature T_c. Such a response is distinct from a uniform heating of the film, for which the peak response is closer to the derivative of the resistance. These observations are consistent with the optical generation of a nonequilibrium distribution of quasiparticles or vortices which can be described by an effective temperature $T^* > T$, where T is the lattice temperature.[6] It is of interest to understand how this mechanism is modified for photon energies near the superconducting gap energy of the metallic grains. In addition, for near and below-gap light the nonequilibrium processes should compete with direct processes, where the intergrain Josephson currents are modulated by the far-infrared radiation. These direct processes have been investigated extensively for point-contact tunneling junctions,[7] microbridges,[8,9] and various types of films. Of particular interest are the microwave response measurements on Sn films by Rose and co-workers,[10,11] and the optical measurements on PbBi oxide films by Enomoto and Murakami.[12] However, none of these various materials exhibited the 2D photoresponse for above-gap light observed for the NbN/BN film.[5] Such a response may have been expected from the PbBi oxide films,[12] but no data to this effect have been presented.

In this paper we examine the photoresponse of a 2D granular film of NbN/BN using far-infrared light in the 2.50 meV (20.2 cm^{-1}) to 18.8 meV (152 cm^{-1}) spectral range. The present measurements extend our previous[13] report of the far-infrared detector responsivity of this same film at 26.0 cm^{-1} at 4.2 K. Our principal observation is that the PR varies more slowly with temperature (for $T < T_c$) for photon energies comparable to or less than the gap energy of NbN ($2\Delta \sim 35$ cm^{-1}). We also show that the dependence of the PR on the light intensity is significantly different for below and above-gap light. The strong dependence of the PR on the magnitude of the superconducting gap is in contrast to the lack of gap-related features in the transmission data for these films. These observations are of interest for high-T_c superconducting films,[14] for which the identification of the superconducting energy gap with standard transmission or reflectivity techniques has been controversial.[15]

Granular NbN/BN films were deposited on sapphire substrates by reactive radio frequency (rf) cosputtering of Nb and BN targets in a nitrogen-argon atmosphere.[2] The film sheet resistance and grain size were dependent on the distance and position of the target relative to the film. Typical films were 5–20 nm thick with room temperature sheet resistances of ~ 1 kΩ/square. Transmission electron microscopy of films comparable to the film studied here shows that the NbN grains have diameters of 5–10 nm, and the BN insulating regions surrounding the NbN grains are ~ 1 nm thick. These films can therefore be modeled as two-dimensional arrays of Josephson junctions. Our 10-nm thick film was scribed for four-probe resistance measurements (current path $\sim 3.0 \times 7.5$ mm) and contacted with indium solder. The biasing circuit and other experimental details have been described previously.[13,14,16]

The film resistance versus temperature near T_c is shown in Fig. 1. The resistive onset for a 0.45-mA bias

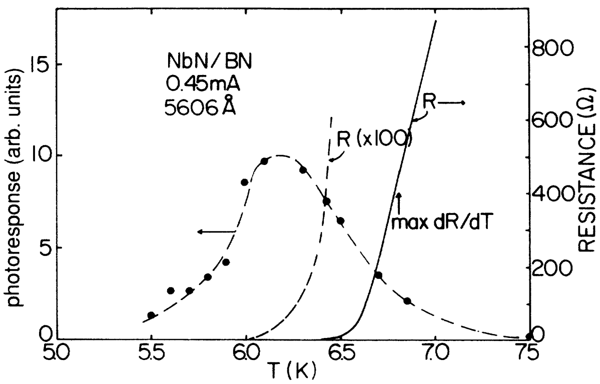

FIG. 1. Photoresponse of NbN/BN film to pulsed light. (560 nm wavelength, 10 ns pulse width, 2μJ/pulse). Resistance of film measured with 0.45 mA bias current.

current is near 6.0 K. The film resistance is found to peak near 15 K at $\sim 1.8k\Omega$, which corresponds to $\sim 700\Omega$/square. The optical response of the film (i.e., change in light-induced voltage drop across the film) as a function of film temperature is also shown in Fig. 1. The radiation source is a pulsed laser operating at 560.6 nm with a 2-μJ pulse energy, a 10-ns pulse width, and a 20-Hz repetition rate. It is evident that the optically induced resistance of the film peaks near the onset of the resistive transition. A response peak near $R=0$ has been predicted by Farrell[17] on the basis of a light-induced reduction of the superconducting gap using the resistively shunted junction (RSJ) model[18] with a thermal-fluctuation averaged solution.[19] The model assumes a low capacitance weak-link Josephson type structure, very much like the films investigated here. This approach yields an average thermally induced voltage given by[17]

$$\langle V \rangle = 2 i_c R \sinh(\beta\pi/2)/\gamma\pi |I_{j\beta}(\gamma/2)|^2 ,$$

where

$$\gamma = h i_c / 2\pi e k T , \beta = h i / 2\pi e k T ,$$

i_c is the critical current, i is the bias current, R is the normal resistance, T is the temperature, and $I_{j\beta}$ is the modified Bessel function with imaginary index. A nonequilibrium response due to a photoinduced increase in quasiparticle density and consequently reduction in the superconducting energy gap 2Δ, is expressed[17] by the chain rule

$$\partial\langle V \rangle/\partial P = (\partial\langle V \rangle/\partial i_c)(\partial i_c/\partial\Delta)(\partial\Delta/\partial P) ,$$

where the first two terms lead to a bolometric response, with a peak near dR/dT. However, for the last term Farrell[17] proposes an expression derived by Parker:[20]

$$\partial\Delta/\partial P \sim [A + \Delta k T \exp(-2\Delta/kT)]^{-1/2} ,$$

which will lead to a peak in the response close to the resistive onset $R=0$ (note: the parameter A is generally a function of i). Although this model assumes an electron pair breaking mechanism, other mechanisms such as an optically induced vortex-antivortex depairing process can be incorporated, with the expectation of a photoresponse peak near the resistive onset.

The far-infrared transmission of the film at temperatures of 6 K (superconducting state) and 20 K (normal state) was measured with a Fourier transform spectrometer. For a homogeneous metal film in the dirty limit, the normal-state transmission is independent of frequency, while the ratio of the superconducting state transmission to normal-state transmission (T_S/T_N) displays[21] a distinct peak at or near the superconducting energy gap frequency (~ 35 cm^{-1} for bulk NbN). The measured normal-state transmission for the NbN film [Fig. 2(a)] is consistent with a conductivity that increases superlinearly with frequency. The measured T_S/T_N ratio (Fig. 3) shows no peak, but instead a very broad minimum indicating increased absorption for the superconducting state. Though an energy gap cannot be determined from these data, the measurements attest to the granular nature of the film.

There have been two earlier studies of the far-infrared properties of granular superconducting films near percolation. Karecki *et al.*[21] observed below-gap absorption in anodized granular NbN films. This absorption increased as frequency decreased in a fashion similar to that calculated by Garner and Stroud[22] using the effective-medium approximation (EMA) for a superconductor-normal metal (*S-N*) mixture. That such films behave as *S-N* mixtures was consistent with the anodizing process used to control granularity which produces conducting oxides at the grain boundaries. In the other work, Carr *et al.*[23] observed in granular lead films a broad absorption centered below the energy gap frequency. The lead grains were separated by insulating silicon oxide. The broad absorption has been interpreted[24] in terms of the EMA for a superconductor-insulator (*S-I*)

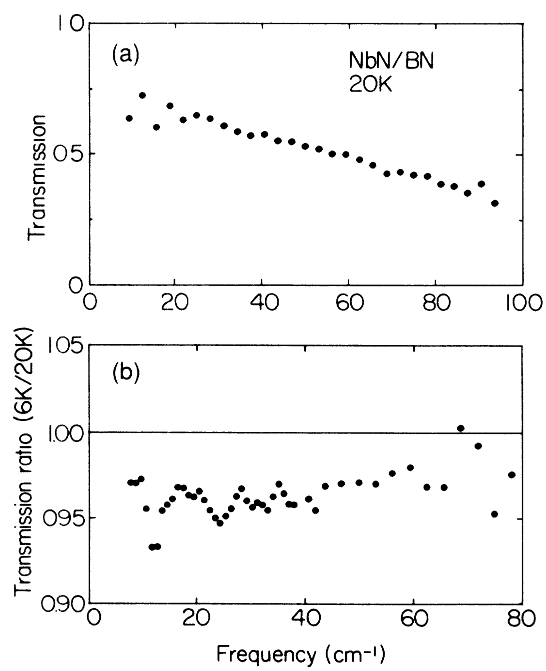

FIG. 2. (a) Far-infrared transmission of granular NbN/BN film in normal state (20 K). (b) Ratio of transmission in superconducting state (6 K) to normal state (20 K) for NbN/BN film.

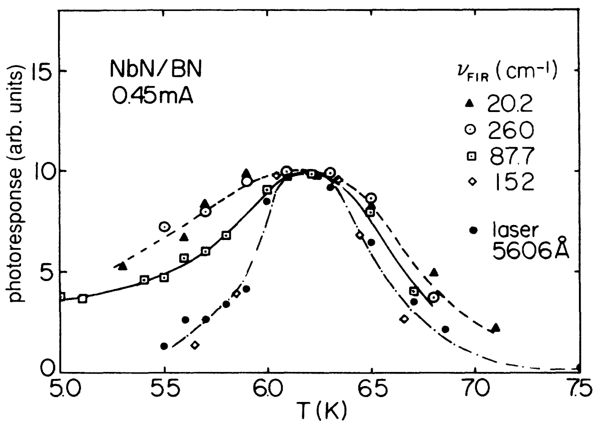

FIG. 3. Far-infrared photoresponse normalized to peak value (near $T \sim T_c$) as function of photon energy.

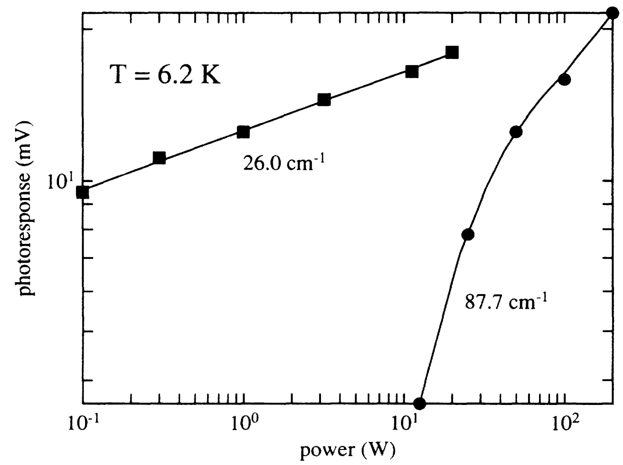

FIG. 4. Dependence of photoresponse on incident power for photons having energies below (26.0 cm^{-1}) and above (87.7 cm^{-1}) the superconducting energy gap of bulk NbN.

mixture very close to conductive percolation. The T_S/T_N ratio for the NbN/BN film more closely resembles that for the granular lead film. Additionally, the normal state transmission for the NbN/BN film is like that of a metal mixture just below (but very close to) percolation.

The photoresponse of the film to far-infrared light was measured with a pulsed superradiant light source[13] which provided 50 ns long pulses of nearly monochromatic radiation at 20.2 cm^{-1}, 26.0 cm^{-1}, 87.7 cm^{-1}, and 152 cm^{-1}. The maximum incident light energy ($\sim 1\mu$J/pulse) was comparable to the maximum 2μJ/pulse of the 560.6-nm laser radiation. In Fig. 3 we show the temperature dependence of the PR for far-infrared radiation and for visible light. At these power levels the rise time[13] is comparable to the rise times of the respective light pulses. The fall time of the response is comparable to the rise time, except that for $T > T_c$ a slow component appears[5] which is attributed to heating effects. This slow component has not been included in the data of Fig. 3. Each experimental point represents an average of the integrated time response (0–50 ns for the far infrared, 0–10 ns for the visible light source). In Fig. 3 we observe that the temperature dependence at 152 cm^{-1} is essentially the same as that measured for visible 5606 Å laser light, but that the far-infrared PR depends less on temperature with decreasing light energy. We have also observed (see Fig. 4) that the PR at 26.0 cm^{-1} is only weakly dependent on far-infrared radiation power over a wide power range from 0.2 to 20 W (0.01 to 1μJ), whereas the 87.7 cm^{-1} response is proportional to power for $P < 30$ W. The observed temperature dependence of the PR as a function of far-infrared wavelength is not sensitively dependent on light intensity over the range of power levels shown in Fig. 4. Also, the distinctly different power dependences observed (see Fig. 4) for 26.0 and 87.7 cm^{-1} are not sensitively dependent on the temperature. Thus, very similar results as those shown in Fig. 4 are observed for $T = 5.5$ K, with the additional observation of a square-law response (PR $\sim P^{1.0}$) for the 26.0 cm^{-1} data at the very lowest power levels (below 0.1 W).

Our observations at 20.2 and 26.0 cm^{-1} are consistent with the direct coupling of far-infrared light to the superconducting weak links in the film. Within the context of the RSJ model[18,25] the time dependence of the phase ϕ across a weak link is obtained from the equation[18]

$$i_{rf}\sin\Omega\tau + i = i_c(d\phi/d\tau + \sin\phi) ,$$

where i_{rf} represents the light-induced current in the junction and where $\tau = (2eRi_c)t/\hbar$, and $\Omega = \hbar\omega/2eRi_c$ are dimensionless variables in time and frequency, respectively. For high-quality semiconductor-insulator-semiconductor (SIS) junctions (our NbN/BN film is expected[2] to consist of a distribution of low-capacitance, high-quality, SIS junctions) the i_cR product can achieve its maximum value $\pi\Delta/2e$ for $T \ll T_{c0}$. With $2\Delta = 35$ cm^{-1}, $\Omega = \hbar\omega/\pi\Delta \sim 0.5$ for the 26.0 cm^{-1} light, and $\Omega = 1.6$ for the 87.7 cm^{-1} light.[26] Russer's analog solution[27] to the above RSJ equation determined that the light-induced change of the dc Josephson current depended critically on the magnitude of Ω. For $\Omega \ll 1$ the light-induced reduction in the zero voltage current has a strong, non-Bessel function dependence on light intensity. In fact, if $\Omega = 0.16$ then the zero voltage response is reduced to 0.1 of its maximum value for $i_{rf} = i_c$, while for $\Omega = 0.64$ this same sized reduction requires that $i_{rf} = 1.8i_c$. Response oscillations observed for single junctions whenever $i_{rf} \gg i_c$ have not been observed for films. Consequently, the PR is not expected to be strongly dependent on rf power or temperature for $i_{rf} > i_c$. An estimate for i_{rf} requires knowledge of the actual absorbed power and the impedance of the film. For anodized NbN films of comparable thickness the far-infrared absorption has been estimated to be $\sim 1\%$ of the incident power. A far-infrared impedance $\sim 100\ \Omega$ for these anodized films could be estimated from the film resistance just above the critical current. For an incident 1 W of far infrared radiation these numbers yield an estimate of $i_{rf} = 10$ mA. The peak in the visible light PR corresponds very close to the critical current condition for this film. Thus $i_c \sim 0.5$ mA at 6.0 K, which suggests that for the present far-infrared powers $i_{rf} > i_c$. The observed "saturation" of the 26.0

cm^{-1} PR is in qualitative agreement with these estimates. The very weak power dependence over two decades in incident power is somewhat surprising. A linear response (PR approximately equal to $P^{1/2}$) has been observed by Rose and co-workers[10,11] for Sn films at microwave frequencies. It is likely that such a $P^{0.5}$ response is obscured by heating effects (evidenced by a long-time component in the time-resolved PR) for power levels greater than 1 W for the 26.0 cm^{-1} data. However, there is no evidence for such heating effects below ~ 1 W of incident power, which suggests that the response mechanism is different for the present film differs from that in the Sn films studied by Rose and co-workers.

For $\Omega \gg 1$ the light-induced change in the zero voltage Josephson current is predicted to have a Bessel function dependence on light intensity.[18,25,27] In addition, the Josephson current is reduced only for large values of i_{rf}/i_c. For example,[25] if $\Omega = 1.67$ the Josephson current is not significantly reduced for $i_{rf}/i_c = 1$. For low rf power ($i_{rf} < i_c$) the response is expected to be square law, as has been verified in detail in the microwave range for single Josephson junctions and granular films. The square-law response is proportional to $i_{rf}^2 i_c R_d$, where R_d is the dynamic resistance (dV/di), which for $T \sim T_c$ will dominate the temperature dependence of the PR in the low power regime.

The measured PR at 87.7 cm^{-1} displays the predicted features of an above-gap direct response. These features include a stronger T dependence than seen for the $\Omega < 1$ data (26.0 cm^{-1}), and a square-law behavior. The observation of a square-law response strongly suggests that the power directly absorbed by the junctions is considerably reduced at 87.7 cm^{-1} compared to 26.0 cm^{-1}. In addition, the magnitude of the direct response is expected to be reduced by a factor $\sim 1/\Omega^2 \sim 1/\omega^2$ for $\Omega \gg 1$. This reduction with increasing frequency is due to the inductive character of the model junction which leads to a shunting of the rf current through the resistive RSJ model circuit element, rather than through the pair current element. The reduction of the direct response with increasing frequency is clearly observed in Fig. 4. As pointed out earlier, a square-law PR is observed at 26.0 cm^{-1} at 5.5 K below 0.1 W of far infrared power. If we extrapolate the linear PR at 87.7 cm^{-1} to 0.1 W, we find the ratio PR(26.0 cm^{-1}) to PR(87.7 cm^{-1})$\sim$200. We note that the above measurements at 26.0 cm^{-1} and 87.7 cm^{-1} span the crossover frequency $\omega_c \sim 55$ cm^{-1}, which is defined by the condition $\Omega = \hbar\omega_c/\pi\Delta = 1$ with $2\Delta = 35$ cm^{-1}.

The dependence of the PR on light energy is further displayed in Fig. 5. Here we have plotted the width of the PR at half maximum in units of K, versus the photon energy. From the RSJ model we expect that the magnitude of the PR due to direct coupling to the Josephson junctions will decrease with increasing light energy as $1/\Omega^2$. We observe in Fig. 5 that such dependence also appears to apply to the temperature dependence of the PR, primarily for $T < T_c$. The generality of this statement must await more detailed measurements of the far-infrared PR of other granular films. [Note that the solid curve in Fig. 5 is described by: $1.55/\Omega^2 + 0.6$ in units of K, where $\Omega = 55$ cm^{-1}/ω (cm^{-1}) and $\Omega \geq 1$]. The results of Figs. 3 and 5 do suggest that any measurements of the frequency dependence of the PR of granular films, for light energies comparable to but greater than the superconducting gap energy, must be carried out as a function of temperature for $T < T_c$.

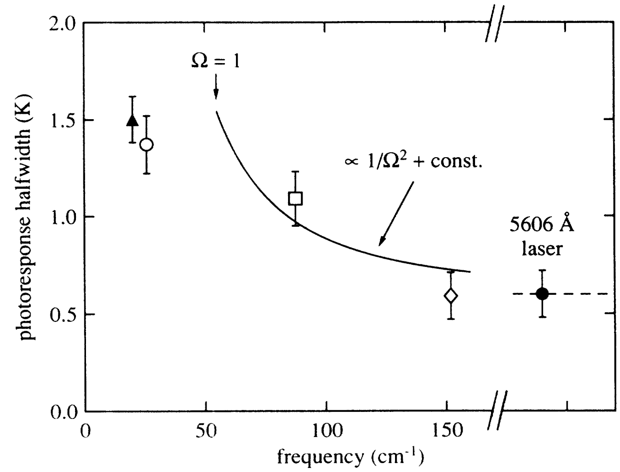

FIG. 5. Width of photoresponse (full width at half maximum) of the curves shown in Fig. 3, as a function of the incident light energy.

We have interpreted our far-infrared data in terms of direct optical coupling to Josephson currents. There may be an alternative, but related explanation. We have previously concluded that the $\hbar\omega \gg 2\Delta$ response is due to the photoresistive processes induced near the 2D transition. This resistance onset has been modeled in terms of depairing of vortex-antivortex pairs. The question may be posed whether photo-induced depairing (which may be direct or through an intermediate thermal process) will be modified as the exciting light energy is lowered through the gap energy of the superconducting grains.[2] It is plausible that for below-gap light the large rf currents channeled through the Josephson element may lead to enhanced vortex depairing and hence resistance induced by vortex motion.

In summary, the far-infrared photoresistive response of a quasi-2D granular NbN/BN film near its resistive onset exhibits a dependence on temperature and light intensity which is a strong function of whether the far-infrared photon energy is greater or less than the superconducting gap of NbN. The results have been shown to be consistent with direct coupling to intergrain Josephson currents for below-gap light, with depairing processes for above-gap light. The observed strong dependence of the infrared PR on the superconducting gap of the grains within the film is likely to be useful for an optical determination of the gap of quasi-2D films of high-T_c superconductors such as $YBa_2Cu_3O_7$.

ACKNOWLEDGMENTS

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