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Optically induced changes in the magnetic properties of the ceramic superconductor $La_{1.8}Ba_{0.2}CuO_4$

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Optically induced changes have been observed in the magnetic properties of the ceramic superconductor $La_{1.8}Ba_{0.2}CuO_4$. The most prominent feature of our data is the light-induced change of the magnetic moment, proportional to $T^{-1/2}\exp(a/kT)$, where a is a constant when the sample is cooled below $\sim T_c/2$. Results obtained in applied magnetic fields of 1, 9, and 26 Oe suggest that the observed effect has contributions from a weakening of the intergranular Josephson junctions under illumination as well as from an additional form of optically induced flux creep, which depends on the applied magnetic field.

I. INTRODUCTION

In experiments by Testardi,[1] destruction of superconductivity in thin films by laser light was demonstrated. He showed that this phenomenon could not be explained simply by heating the sample with light, but rather with a generation of excess unpaired electrons. Following this idea Owen and Scalapino[2] furnished a theory of nonequilibrium superconductors predicting a reduction of the energy-gap parameter, $\Delta(T)$, by a pair-breaking mechanism such as a flux of photons. The results of this model and those of a more rigorous theory[3] agree with the experimental data of Parker and Williams[4] for tunnel junctions.

Nonbolometric optical response of films of high-T_c oxide superconductors has been reported by several authors. Leung *et al.*[5] attributed their results to photoinduced phase slips and Zeldov *et al.*[6] to optically induced flux creep. Iwasaki[7] has used the idea of light-induced breaking of Cooper pairs to discuss the response of optical detectors made of high-T_c oxide materials. Recently, Culbertson *et al.*[8] have reported an observation of nonequilibrium heating in Y-Ba-Cu-O films.

In comparison with metallic films, high-T_c superconductors have a lower reflection coefficient of light. This feature makes them attractive for investigations of the influence of light on their superconducting properties. Ceramic oxide superconductors consist of grains connected to each other by weak superconductor-insulator-superconductor-type links.[9,10] When an external magnetic field is applied, closed loops of screening currents will be generated both around the edges of the grains and through junctions between the grains. The possible paths of the supercurrent in the sample depend on the critical currents of the links.[11] It is expected that any changes induced by light in the properties of intergranular junctions or in flux trapping forces will influence the magnetic moment of the sample.

We will report here an investigation of optically induced changes of the magnetic properties of the ceramic superconductor $La_{1.8}Ba_{0.2}CuO_4$. Measurements were made in external fields where the magnetic behavior of the sample is governed mainly by the Josephson weak links or by the London surface currents, respectively.

II. EXPERIMENT

The sample used in the experiments is ceramic $La_{1.8}Ba_{0.2}CuO_4$ prepared from the primary materials by successive grinding, heat-treatment, and sintering processes. The superconducting transition temperature was found to be $T_c = 36$ K as determined by a conventional superconducting quantum interference device (SQUID) magnetometer. To detect light-induced changes in the magnetization of the sample it was inserted in a magnetometer provided with optical access.

The principle of the used magnetometer is shown in Fig. 1. The sample is illuminated by a tungsten lamp from the top of a dewar with an optical fiber. The tube containing the fiber ends with an extension made out of fused silica to accommodate the sample holder, heater, and a carbon-glass thermometer. On the outer wall of the silica tube a ten-turn detection coil is wound (diameter 10 mm) matched to the input of an rf SQUID. This tube is fixed tightly to a superconducting magnet and the whole assembly is shielded against stray magnetic fields with superconducting and μ-metal shields. The magnetization signal is detected with a lock-in amplifier connected to the output of the SQUID control electronics. To separate the optically induced signal from any slow drifts

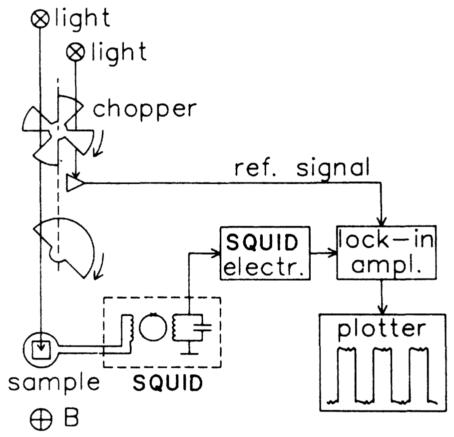

FIG. 1. Experimental setup used for investigations into photoinduced changes of the magnetization.

of the background, the excitation light is chopped both at a high frequency (67 Hz) and a low frequency (180° plate in Fig. 1; 1 rev/min). The setup is calibrated with a circular test coil having the same diameter as the sample disc (3 mm). The measurements could be done between 2–50 K with the sample tube either evacuated or filled with He exchange gas.

III. EXPERIMENTAL RESULTS

We have observed optically induced change of magnetization, M_{opt}, in the $La_{1.8}Ba_{0.2}CuO_4$ sample and investigated the dependence of this effect on temperature, applied magnetic field, intensity of light, and the way of cooling the sample. When superconducting metallic films are illuminated by laser-light heating of the sample is a problem in experiments aiming to detection of light-induced nonequilibrium phenomena.[1] To exclude the possibility that the effect observed in the present work could result simply from change of the magnetization by heating when the sample is exposed to light we measured the increase of its temperature under continuous illumination. The temperature rise was found to be less than 80 mK in the range of pumping-power densities 0.1–1.5 mW/mm^2. As shown in Fig. 2, M_{opt} depends linearly on pumping power between these limits. On the other hand, the chopped light used in the measurements consisted of 7.5-ms long pulses separated by dark periods of the same length. To calculate the corresponding temperature rise we assume that pumping power is 1 mW or 7.5×10^{-6} J. Using the value of the heat capacity $C=70$ mJ/mol K (measured for $La_{0.8}Ba_{0.2}CuO_4$ at 6 K)[12] and the thermal conductivity $K=0.7$ W/m K (determined for ceramic $YBa_2Cu_3O_6$)[13] the temperature rise of the sample (mass 55 mg) turns out to be 0.13 K.

It can be concluded from the existing data that C is proportional to T^3 (Ref. 12) and that K probably decreases by a factor of 10 when the temperature of the sample decreases from 20 to 5 K. According to a simple thermal calculation for our uniformly illuminated disc-shaped samples (diameter about 3 mm and thickness 0.5 mm), a much more pronounced upturn of the change of magnetization would result from heating by light at low temperatures than was observed for M_{opt}. This suggests that the thermal mechanism based on uniform lattice heating cannot explain our results. More evidence against this mechanism is obtained by careful magnetic measurements.

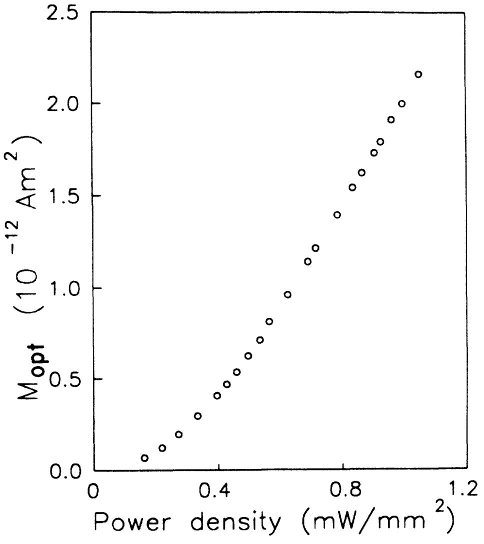

FIG. 2. Optically induced change of the magnetic moment M_{opt} in $La_{1.8}Ba_{0.2}CuO_4$ vs pumping-power density.

Figure 3 shows the temperature dependence of the magnetization of the sample measured with a conventional SQUID magnetometer when it is cooled in zero external field (ZFC) or in a field (FC). These two measure-

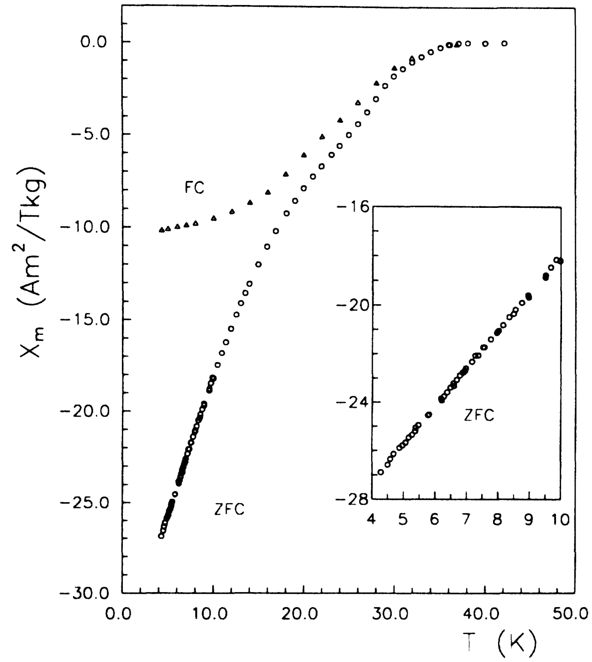

FIG. 3. Temperature dependence of the magnetization of $La_{1.8}Ba_{0.2}CuO_4$ determined in the field of 5 Oe after cooling the sample in zero external field (ZFC) or in the measuring field (FC) (without optical excitation).

ments are related to the shielding effect (ZFC) and to the Meissner-effect (FC), respectively. For $T < 12$ K the ZFC magnetization shows a linear increase of 1.6×10^{-13} $\mathrm{A\,m^2\,K^{-1}\,mg^{-1}\,Oe^{-1}}$ when the temperature of the sample decreases. For the FC magnetization (the sample was cooled in a field of 5 G) the corresponding increase is only about 2×10^{-14} $\mathrm{A\,m^2\,K^{-1}\,mg^{-1}\,Oe^{-1}}$. Temperature dependence of the observed photomagnetic effect is shown in Fig. 4. Assuming that it arises from the change of the magnetization of the sample due to heating by light, the ZFC and the FC plots of M_{opt} should resemble those shown in Fig. 3. This is not, however, the case as the temperature derivatives $\partial M/\partial T$ and $\partial M_{\mathrm{opt}}/\partial T$ differ clearly.

Due to strong absorption the light can penetrate only into a thin surface layer of the sample. In this situation optical radiation may increase the number of phonons with energy greater than twice the superconducting energy gap. Parker[14] has suggested that the high-energy phonons will heat the quasiparticles to an effective temperature T^* higher than the temperature at which the electrons and the lattice are in thermal equilibrium. He has also shown that optically induced nonequilibrium properties of metallic film superconductors can be interpreted equally well with this "modified heating model" than with the Owen-Scalapino theory.[2,14] It is likely that the nonequilibrium heating mechanism is behind the photomagnetic effect observed in this work. As a result of nonequilibrium heating the superconductor will have properties similar to those of a BCS superconductor at the temperature T^*. From comparison between Figs. 3 and 4 it is interesting to find that the temperature dependence of M_{opt} observed below 15 K has the shape which is expected from ordinary magnetization measurements above 25 K (where the FC and the ZFC magnetizations become equal). The value of T^* about 20 K above the ambient temperature is not unrealistic.

The most noticeable feature of the light-induced change of the magnetization of the sample is the maximum observed around 5 K. As shown in Fig. 4(a) the temperature dependence of M_{opt} can be fitted above 8 K with a function

$$M_{\mathrm{opt}} \propto T^{-1/2} \exp(a/kT) \;, \tag{1}$$

where $a = 15.2$ K. Our measurements were made in external fields of 1, 9, and 26 Oe, selected on the basis of the magnetic properties of the sample as will be discussed later. Comparison of the curves in Figs. 4(a)–4(c) reveals a difference between the curves measured with the sample cooled in zero external field or in the field, respectively. When the measurement is made in the field of 1 Oe this difference is visible only below 6 K, but in higher fields it can be seen up to about 15 K. At the maximum of the plots of M_{opt}, a small anomaly is observed. This feature is more clearly seen for ZFC samples and its relative strength depends on the magnetic field (Fig. 5).

Light-induced changes of the magnetization were

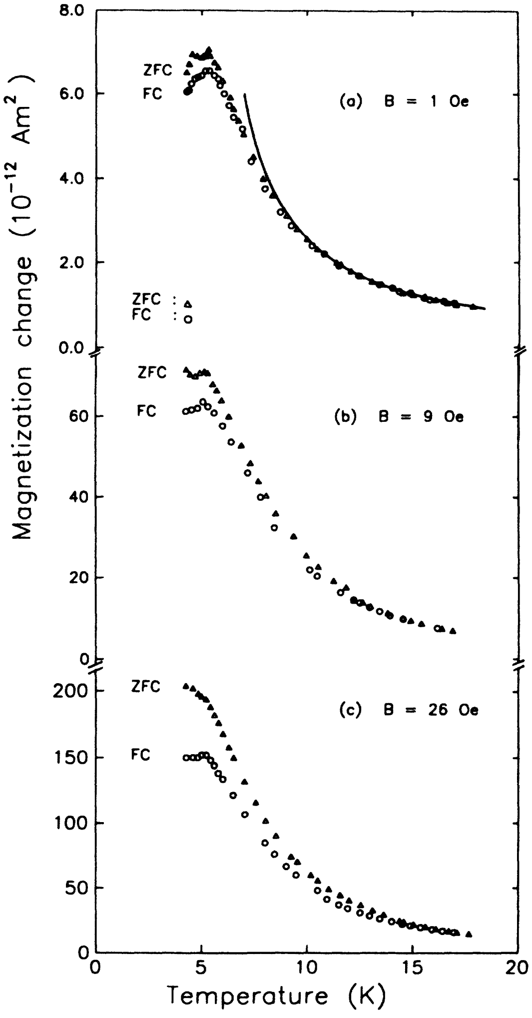

FIG. 4. Temperature dependence of M_{opt} measured in fields of 1, 9, and 26 Oe. The solid line in (a) represents a fit to Eq. (1).

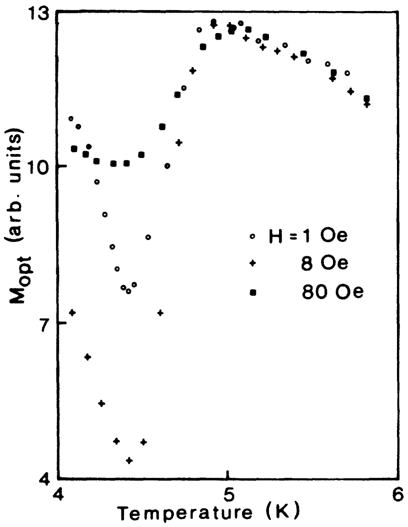

FIG. 5. Magnetic-field dependence of the anomaly in M_{opt} observed between 4 and 5 K. The data points obtained in different magnetic fields are scaled against the maximum of M_{opt} observed in these fields.

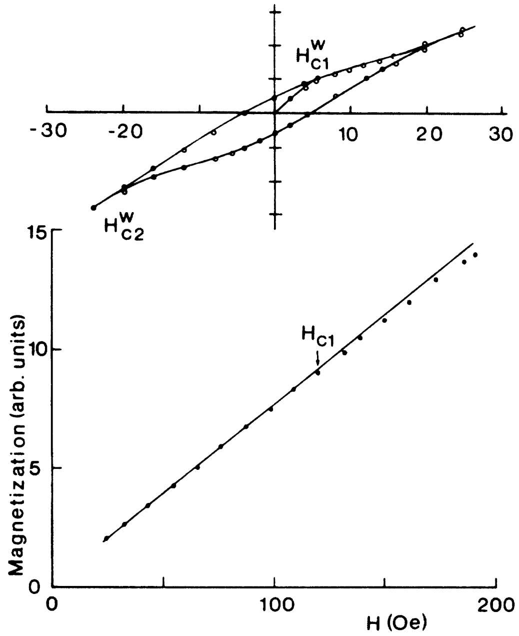

FIG. 6. Low-field magnetization measurements used to characterize the Josephson hysteresis loop and to estimate the first critical field H_{c1} of the sample.

found to be sensitive to the applied magnetic field. For this reason low-field magnetic properties of the sample were investigated with the conventional SQUID magnetometer. In the range of lowest fields a hysteresis loop is observed. As can be seen from the plot in Fig. 6 the end point of the initial curve of this loop is $H_{c1}^{w} \simeq 4$ Oe and the closing point is $H_{c2}^{w} \simeq 20$ Oe. In higher fields the plot of M versus H reveals a point above which $M(H)$ starts to depart from linear behavior. This point can be defined as the first critical field or $H_{c1} \simeq 124$ Oe in our sample. The existence of these three critical fields is common in ceramic superconductors.[15] The low-field-hysteresis loop is attributable to Josephson junctions for $H < H_{c1}^{w}$. Between H_{c1}^{w} and H_{c2}^{w} the junction behaves like a type-II superconductor with its own penetration depth λ_j, critical fields (H_{c1}^{w} and H_{c2}^{w}), and screening current flowing at the edge of the junction in the penetration layer.[16] Beyond H_{c2}^{w} most of the junctions have been broken. In the range of $H_{c2}^{w} < H < H_{c1}$ the magnetic properties are governed by London surface currents around the grains and in the range of $H > H_{c1}$ by vortex pinning within the grains.

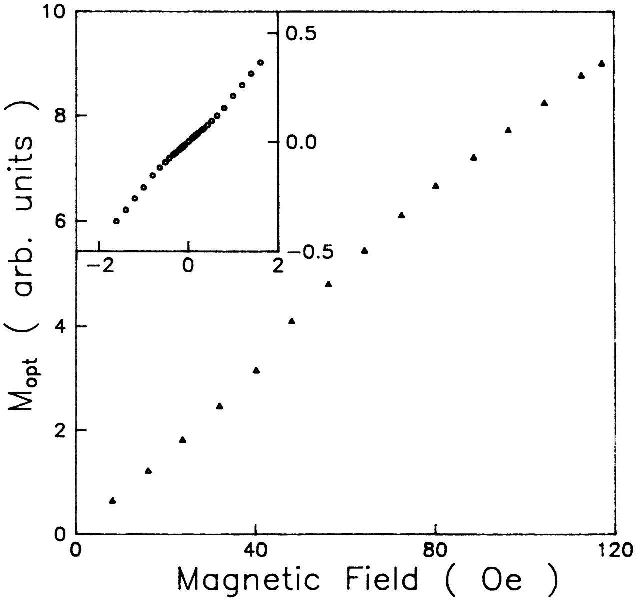

FIG. 7. Magnetic-field dependence of the optically induced magnetic moment.

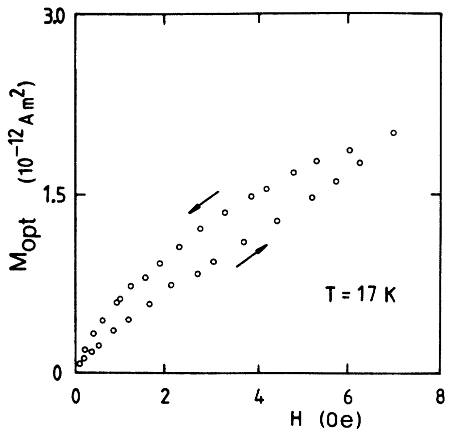

FIG. 8. Hysteresis of M_{opt} when the field is cycled.

Figure 7 displays the dependence of M_{opt} on the applied magnetic field. It is found that when the direction of the field is reversed, the sign of M_{opt} is changed (inset of Fig. 7). Two anomalies are observed with increasing field. The first one takes place around $H = 1$ Oe. The other anomaly is found at about 40 Oe and can be related with a crossover region to a state where the material is expected to behave like a collection of independent filaments of radius equal to the grain size.[15]

An example of the plot of M_{opt} versus magnetic field is shown in Fig. 8 when the field is cycled up and down. We attribute the observed irreversibility of M_{opt} to trapping of flux in the sample. When the applied field increases, local screening currents are induced and arranged in a way determined by flux-pinning forces. On decreasing the field part of the flux is trapped in the intergranular links and other pinning centers leading to the observed cycle of M_{opt}. The amount of trapped flux was found to increase when the extent of the field sweep was increased. This feature, as well as the location of H_{c1}^{W} in the low-field-hysteresis loop (Fig. 6), resemble the nonreversible properties of microwave absorption observed in various field sweeps for $\mathrm{La_{1.85}Ba_{0.15}CuO_4}$.[17] The appearance of nonresonant microwave absorption in the sample was associated with flux slips in a weakly random system.[17] Therefore, it is closely linked with the irreversibility of M_{opt} found in our experiments.

IV. DISCUSSION

In our experiments the $La_{1.8}Ba_{0.2}CuO_4$ sample was excited at wavelengths corresponding to energies between 1 and 3 eV. Because of strong absorption, light can penetrate into a layer representing for only a fraction of the grain diameter. The SQUID output signal ($\propto M_{\text{opt}}$) shown in Fig. 2 is at small intensities at nonlinear and at higher intensities a linear function of the pumping power. It is likely that the nonlinear part is caused by a thin surface layer which has properties differing from those of the interior of the grains.

The most prominent change of $M_{\text{opt}}(T)$ was observed below 10 K. It is expected that in this region ($T<T_c$) the superconducting gap is practically independent of temperature[18] and the grains of the bulk ceramic sample are extensively phase locked.[19,20] When a small external magnetic field is applied part of the weakest intergranular junctions are destroyed and only those which can carry the critical current needed for shielding out the external flux are left. The supercurrent flowing through a junction connecting grains i and j is given by[20]

$$I_{ij}=I_c(T)\sin(\Phi_i-\Phi_j-A_{ij}) , \tag{2}$$

where A_{ij} is the gauge-invariant phase factor

$$A_{ij}=\frac{2\pi}{\Phi_0}\int_i^j \mathbf{A}\cdot d\mathbf{l} \tag{3}$$

and $\mathbf{A}$ is the vector potential of the field. The integral is taken along the line connecting the grains i and j. The critical current density of the junctions can be given by the Ambegaokar-Baratoff equation[11]

$$I_c(T)=\frac{\pi\Delta(T)}{2eR_{nn}}\tanh\left[\frac{\Delta(T)}{2kT_c}\right] , \tag{4}$$

where R_{nn} is the resistance of the junction in the normal state.

Ebner and Stroud[21] have presented a theory of diamagnetic susceptibility of weakly linked two-dimensional clusters of superconducting grains. An important idea in their model is that at finite magnetic fields a cluster with closed current loops cannot find a state which simultaneously minimizes all the bond energies. This "frustration" is caused by the phase factors A_{ij} which make some of the bonds between grains ferromagnetic (phases Φ_i and Φ_j are equal) and some antiferromagnetic or favoring an angle between phases. A frustrated cluster with many closed loops has a number of ground states with nearly equal energy. When the external field is varied the cluster executes hops from one configuration to another to stay in the state of the minimum energy. This is possible as long as the cluster is in thermal equilibrium. It was suggested[21] however, that at low temperatures the cluster may be trapped in metastable configurations. Then it is possible to reach the ground state only if sufficient energy is available to cross the barrier hindering the transition. Using their model Ebner and Stroud were able to calculate the magnetic moment of the clusters and showed that they have properties similar to spin glasses.

A bulk sample of ceramic high-T_c superconductor contains a large number of grains and clusters of them having different size and coupling strengths. In addition to Josephson links, between the grains there may exist intragranular junctions and defects which are able to pin the flux. The most prominent feature of the results in Fig. 4 is the nearly exponential increase of the light-induced magnetic moment change when the temperature of the sample is decreased. The part of the sample in which the light can penetrate is an approximately two-dimensional sheet on its surface. Supposing that I_{ij} is the Josephson current from grain i to grain j, the magnetization of a cluster of grains situated at $\mathbf{R}$, far from an observer, is

$$\mathbf{M}=\sum_{\langle ij\rangle} I_{ij}\mathbf{r}_{ij}\times\mathbf{R}/R^3 , \tag{5}$$

where $\mathbf{r}_{ij}$ is the vector distance from grain i to grain j. Comparing Eqs. (4) and (5) it is found that the magnetization due to current flow in the cluster can be influenced by a perturbation reducing $\Delta(T)$.

The result for the energy gap of a superconductor pumped by light to a nonequilibrium state is for low reduced temperatures[2]

$$(\Delta/\Delta_0)^3=([(\Delta/\Delta_0)^2+n^2]^{1/2}-n)^2 , \tag{6}$$

where Δ is the reduced energy gap, Δ_0 is the unperturbed gap at $T=0$, and n is the excess quasiparticle number density in units of $4N(0)\Delta_0$. Here $N(0)$ is the density of states (for one spin) at the Fermi level. Quasiparticles will be generated at a rate proportional to the optical flux P and to their effective recombination time τ. In a steady state[4]

$$n=\frac{rP\tau}{4N(0)\Delta_0 V} , \tag{7}$$

where r is the number of quasiparticles produced per photon in a unit time and V is the illuminated sample volume. There are two limiting cases in the temperature dependence of τ.[22,23] If the excess number density of quasiparticles $\delta N=4N(0)\Delta_0 n$ is much smaller than the number of thermally excited quasiparticles, N_T, one finds a strong dependence $\tau\sim T^{-1/2}\exp[\Delta(T)/T]$. For the opposite extreme, $\delta N/N_T\gg 1$, τ is independent of T. We can fit the experimental result in Fig. 4(a) with Eq. (1) using $a=\Delta(T)/k=15.2$ K. Hence the value of $\Delta(T)$ obtained from the fit is about 25% from Δ_0 calculated for $T_c=36$ K from the BCS theory. The form of $M_{\text{opt}}(T)$ was found to depend somewhat on the pumping power although it could always be fitted with a function given by Eq. (1). A saturation of $M_{\text{opt}}(T)$ is observed at about 5 K corresponding to $\Delta/kT\simeq 3$. A similar behavior has been reported for optically induced decrease of the energy gap of a Pb-Pb-oxide-Pb junction.[4] Following this work we determine the value of n at a point defined by $\delta N/N_T=1$ ($\sim$7.2 K). Substituting the result $n=0.11$ into[2,14]

$$\frac{\Delta}{\Delta_0}\simeq 1-2n , \tag{8}$$

which is obtained from Eq. (6) at small pumping powers, we get $(\Delta/\Delta_0)\simeq 0.78$.

Although our data agrees qualitatively with the results of Owen and Scalapino,[2] there is a disagreement (by a factor of 2–3) between Δ determined from fitting the data with Eq. (1) or calculating it from the condition $\delta N/N_T = 1$.

It is obvious that the model where the light-induced change of the magnetic moment of the sample is assumed to be directly proportional to $I_c(\Delta)$ is too crude. It neglects the dependence of M_{opt} on the microscopic structure of the sample. This becomes significant when the critical currents are reduced to the extent that in the applied field the shielding currents must find a new configuration between the grains. As proposed by Ebner and Stroud[21] the available metastable configurations depend on temperature. The structure of $M_{\mathrm{opt}}(T)$ at low temperatures (see Figs. 4 and 5) can be understood by optically induced hops of flux from frozen-in metastable configurations.

The ZFC curves in Fig. 4 were obtained by cooling the sample from above T_c to 4 K in zero magnetic field and detecting the magnetization when it was slowly warmed up in the measuring field of 1, 9, or 26 Oe. After that the FC magnetization was measured in the same field during a cooling cycle down to 4 K. The difference between the obtained ZFC and FC curves can be understood by pinning of flux in the sample. During the warming up period, part of the pinned flux is released by thermal and optical excitation. When the sample is cooled down again the contribution of the released flux is missing from the observed M_{opt}. In fields above 1 Oe the difference between the results from the ZFC and FC measurements can probably be attributed to the intragranular flux motion in the presence of the Lorentz force.

Possible mechanisms for optically induced motion of flux comprise of light-induced pair breaking in weak junctions, as discussed above, generation of optical phonons at pinning centers, change of local electronic properties of material by illumination, etc. The effects observed by us are produced in a surface layer of the order of 1 μm thickness when the diffusion of the quasiparticles is taken into account. In ceramic material where flux can move relatively easily at grain boundaries, one can expect photoinduced changes in properties of SQUIDS and other devices based on weak-link Josephson junctions.

V. CONCLUSION

We have observed light-induced change in the magnetic moment of the ceramic high-T_c superconductor $La_{1.8}Ba_{0.2}CuO_4$. Comparison with the temperature dependence of the initial magnetization of the investigated material shows that the observed effect cannot be explained simply by heating the sample with light.

The change of the magnetization by illumination has a nearly exponential temperature dependence down to 8 K, below which $M_{\mathrm{opt}}(T)$ gradually saturates showing some oscillation. This behavior is discussed by using a model in which the sample consists of a network of supercurrent loops controlled by weak-link Josephson junctions. A qualitative agreement with the experimental data can be obtained by assuming that the flux pinned by the junctions is influenced by reducing optically the superconducting energy gap. This is basically the same mechanism, which has been used to explain light-induced nonequilibrium phenomena in tunnel junctions made of classical superconductor films.[1,4] In $La_{1.8}Ba_{0.2}CuO_4$ a quantitative discussion of the data is difficult for several reasons. These involve the uncertainty of the value of Δ_0, the ambiguity of the effective quasiparticle recombination time, which may depend on both temperature and the quasiparticle excitation energy,[23] and the complex structure of our ceramic sample. The data of $M_{\mathrm{opt}}(T,H)$ reveal an additional form of photoinduced flux motion, which depends only weakly on temperature and increases in proportion to the applied magnetic field.

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