

## Nernst effect by laser-pulse heating in Tl-Ba-Ca-Cu-O superconducting thin films

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The Nernst effect has been observed in a high-temperature superconductor with a laser-pulse-heating method. Irradiating a Tl-Ba-Ca-Cu-O film by a short laser pulse, a voltage signal transverse to an external, surface-parallel magnetic field has been observed. This signal is attributed to vortex depinning and vortex transport driven by the laser-induced temperature gradient. We describe our results by thermal flux tube activation and find a distribution of pinning energies from about 100 to 4000 K.

### INTRODUCTION

The dynamics of magnetic flux lines in high- $T_c$  superconductors is of strong current interest. Recently published work on dissipation due to flux-line flow indicates large discrepancies in experimental findings and interpretations. Measurements of the damping of mechanically oscillating  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and  $\text{Bi}_{2.2}\text{Sr}_2\text{Ca}_{0.8}\text{Cu}_2\text{O}_8$  single crystals in a magnetic field yielded large loss peaks at temperatures below  $T_c$ .<sup>1</sup> This effect was attributed to melting of the flux-line lattice. Similar mechanical resonance measurements were carried out applying a vibrating reed technique to polycrystalline Bi-based superconducting samples.<sup>2</sup> The observed mechanical loss was ascribed to flux-line depinning and dissipative flux-line motion. Depending on the superconducting material, depinning temperatures of not more than  $T_c$  were deduced. On the other hand, a magnetic-field-induced resistivity following an Arrhenius law has been attributed to a dissipation mechanism due to thermal-activated flux-line motion.<sup>3,4</sup> In  $\text{Bi}_{2.2}\text{Sr}_2\text{Ca}_{0.8}\text{Cu}_2\text{O}_{8+x}$  crystals at low magnetic field strengths on the order of 0.1 T, activation energies of about 1000–3000 K have been found, depending on the orientation of the magnetic field with respect to the  $(a,b)$  plane and decreasing with rising field strength.<sup>3</sup> As a surprising result, the observed resistance as function of temperature at constant magnetic field could be described by a single activation energy instead of an energy distribution as expected from a variety of pinning strengths. Even larger activation energies up to  $2 \times 10^5$  K have been found in  $\text{YBa}_2\text{Cu}_3\text{O}_7$  single crystals.<sup>4</sup> More recently, the interpretation of resistive current flow due to flux-line depinning and dissipative flux-line motion was questioned by Woo *et al.*<sup>5</sup> The resistance of polycrystalline  $\text{Tl}_2\text{Ba}_2\text{CaCu}_2\text{O}_x$  films was found to be the same for the magnetic field being normal or parallel to the current, indicating that the Lorentz force does not play a significant role for the magnetic-field-induced resistance below  $T_c$ .

In the present article we report on a new method to investigate flux-line motion in high- $T_c$  superconductors. Tl-based thin superconducting films were subjected to an external magnetic field parallel to the surface. Irradiating

a film by a short laser pulse, a strong temperature gradient is obtained perpendicular to the film surface, and at the same time, a voltage signal is observed transverse to the magnetic field and transverse to the temperature gradient. In analogy to the Nernst effect in normal metals and semiconductors, the effect giving rise to this voltage is called Nernst effect in type-II superconductors. A Nernst effect in a conventional type-II superconductor was first observed in 1966,<sup>6</sup> and the effect was interpreted in terms of vortex motion and transport of entropy by the moving vortices.

### EXPERIMENTAL

The measurements were carried out on polycrystalline Tl-Ba-Ca-Cu-O superconducting films of about  $1 \mu\text{m}$  thickness showing transition temperatures near 100 K. According to x-ray diffraction measurements, the films are predominantly of the 2:2:1:2 phase. An x-ray diffraction pattern of a film is shown in Fig. 1. Comparing the experimental pattern with a calculated powder pattern,<sup>7</sup> good agreement in the location and also in the intensity of the diffraction peaks is found, and we conclude that our samples are polycrystalline and essentially not oriented.  $T_c$  values around 100 K are typical for the 2:2:1:2 phase.<sup>7</sup> The film preparation procedures and details on the morphology of the films were described elsewhere.<sup>8,9</sup> The films were deposited on  $10 \times 10 \times 1 \text{ mm}^3$   $\text{SrTiO}_3$  substrates and had bridgelike structures. The experimental arrangement is shown in Fig. 2(a). The magnetic field was generated by a stack of small CoSm solid-state magnets, yielding a field strength of about 0.1 T with the field vector parallel to the plane of the film. Contacts for measuring the transverse voltage were made with silver epoxy paint. The contact regions were shielded by a metallic mask to prevent irradiation of the contacts. The whole arrangement was placed in a temperature variable cryostat with optical access.

The films were irradiated with TEA- $\text{CO}_2$  laser pulses (duration, 100 ns; repetition rate,  $1 \text{ s}^{-1}$ ). The energy of the laser pulses was varied by calibrated attenuators; the maximum pulse energy density at the film surface was about  $20 \text{ mJ/cm}^2$ . Signals were recorded with a fast storage oscilloscope. In Fig. 2(b) two signal pulses are

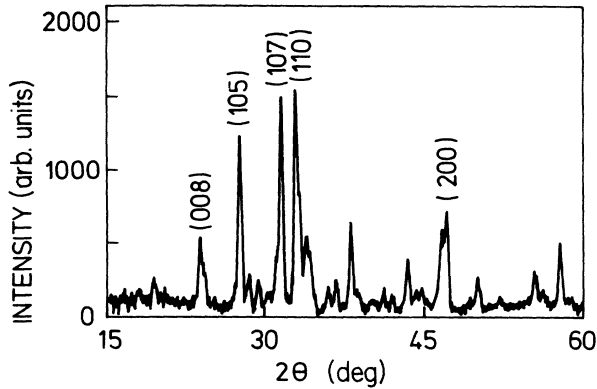


FIG. 1. X-ray diffraction analysis of a TI-Ba-Ca-Cu-O thin film produced by a method combining a laser ablation and a thermal diffusion technique (Ref. 8). The film is predominantly of the 2:2:1:2 phase.

shown, demonstrating the Nernst effect; upon reversing the orientation of the magnetic field, the polarity of the signal voltage becomes reversed.

In Fig. 3 a series of recordings for different laser-pulse energy densities and an initial temperature of the sample of  $T_I = 30$  K is plotted. The time-resolved Nernst signals with maximum voltages up to 1 mV show a fast increase, being as fast as the laser pulse, and a slow decrease of about  $1 \mu\text{s}$ . As the Nernst voltage is proportional to the temperature gradient  $\nabla T$ ,<sup>10</sup> the signals indicate the dynamics of the buildup and decay of a temperature gradient across the high- $T_c$  film. The temperature gradient assumes a maximum at the end of the laser pulse and decays afterwards. With increasing pulse energy, part of the film reaches even final temperatures  $T > T_c$ . In this case we observe, after a first maximum of the Nernst signal, a decrease of the signal due to the film becoming partly normal conducting. The Nernst signal increases again at larger times when the film has cooled down so that  $T < T_c$  everywhere. With further increasing pulse

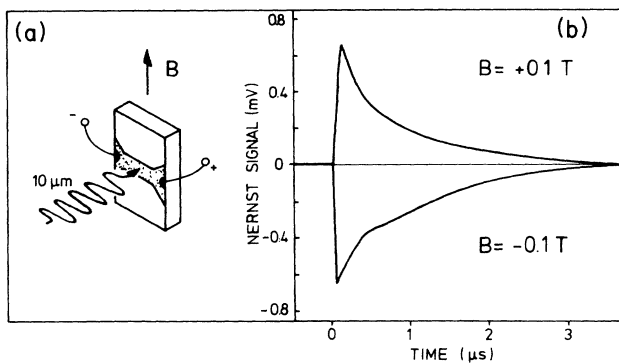


FIG. 2. (a) Experimental arrangement and (b) signal pulses corresponding to the magnetic field  $B$  pointing in opposite directions in the plane of the high- $T_c$  film.  $B > 0$  corresponds to the orientation of  $B$  with respect to the signal polarity shown in (a). Laser-pulse energy density  $E_p = 8 \text{ mJ/cm}^2$ ; initial film temperature  $T_I = 30$  K.

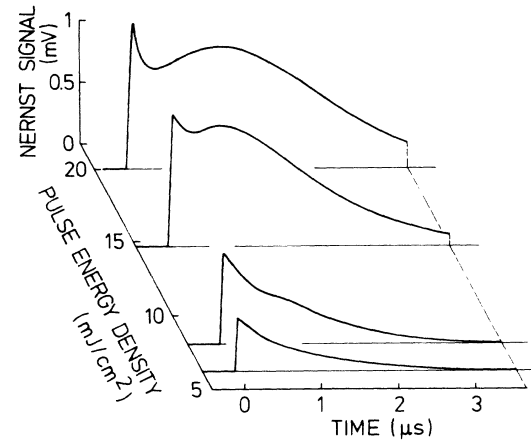


FIG. 3. Nernst signal pulses for various laser-pulse energy densities  $E_p$ ; initial film temperature  $T_I = 30$  K.

energy, the second peak shifts to large times, indicating an increase of cool down time, whereas the signal heights stay approximately constant.

The height of the Nernst voltage versus laser-pulse energy density  $E_p$  is plotted in Fig. 4 for various initial sample temperatures  $T_I$ . The signals show a superlinear onset at low energies, an increase with a slope depending on  $T_I$ , and finally saturate at about the same maximum value for all  $T_I$ 's. The saturation of the signal indicates that for high pulse energies  $T_c$  of the superconducting film is always surpassed. In Fig. 5 the signal height measured at a constant nonsaturating laser-pulse energy of  $3 \text{ mJ/cm}^2$  as a function of  $T_I$  is shown. The signal height is nearly constant at low temperatures, shows a maximum near  $T_c$ , and vanishes above  $T_c$ .

For magnetic fields large compared to the critical field  $H_{c1}$ , where flux penetration into the superconductor sets in, the density of flux tubes is proportional to the applied magnetic field. In this case, the Nernst voltage is expected to be proportional to the applied field. To check this dependence, an experiment using a superconducting magnet instead of the solid-state magnets was performed. The high- $T_c$  film was immersed in liquid helium at 4.2 K,

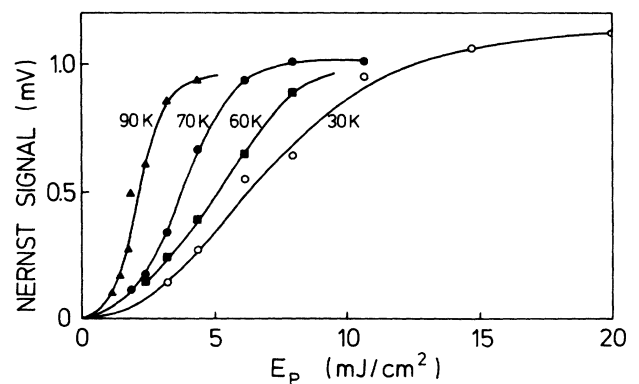


FIG. 4. Signal height vs irradiation energy density  $E_p$ , for various initial film temperatures  $T_I$ .

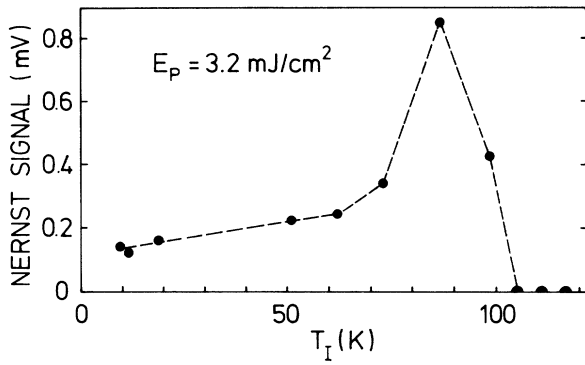


FIG. 5. Signal height for a low laser-pulse energy density of  $E_p = 3 \text{ mJ/cm}^2$  as function of temperature  $T_I$ ; the signal vanishes if the sample becomes normal conducting.

and the laser-pulse energy was kept constant. The Nernst voltage was measured up to a magnetic field of 5 T. Within the experimental error, we find a linear dependence of the signal on magnetic field as shown in Fig. 6. The same dependence, with an inverted Nernst signal, is found for reversed magnetic field.

### DISCUSSION

The Nernst voltage is generated by flux motion between two contact points.<sup>11</sup> The voltage is proportional to the rate at which flux crosses a curve joining the contacts. The rate depends on the fluxon velocity, which is proportional to the thermal force  $f_{\text{th}} = -S\nabla T$ ,<sup>10</sup> where  $S$  is the fluxon transport entropy and  $\nabla T$  is the temperature gradient, and on the depinning probability, i.e., the thermal-activated hopping rate  $\nu = \nu_0 \exp(-T_p/T_F)$  of fluxons. In this expression  $\nu_0$  is the attempt frequency,  $T_p$  is the temperature corresponding to the pinning energy, and  $T_F$  is the film temperature.

Because the fluxons move from the hot (front) film surface to the colder film reverse and because this motion depends upon flux depinning in this colder region of the film, we chose for  $T_F$  a film temperature near the interface between film and substrate. For small laser-pulse en-

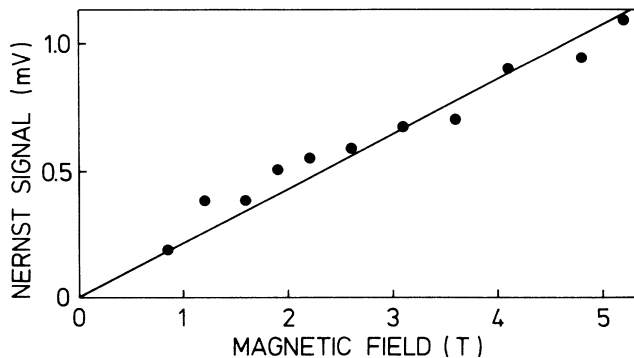


FIG. 6. Signal height vs magnetic field for constant laser-pulse energy. The sample was immersed in liquid helium at 4.2 K for this measurement.

ergies, as long as the variation of the specific heat and thermal diffusivity in the film are negligible, the film temperature rise is proportional to the incident laser-pulse energy density  $E_p$ . The temperature distribution in the film has been calculated by applying an instantaneous plane source model,<sup>12</sup> assuming that 20% of  $E_p$  is absorbed within a penetration depth of 100 nm. These latter values have been derived from spectroscopic measurements.<sup>13</sup> The thermal diffusivity was obtained from bolometric measurements,<sup>14</sup> yielding values very similar to those of sintered Y-Ba-Cu-O samples.<sup>15</sup> Then the temperature difference  $\Delta T$  between the film surfaces and the temperature  $T_F$  may be described as  $\Delta T = (\nabla T)d = A(T_I)E_p$  and  $T_F = B(T_I)E_p + T_I$ , respectively, where  $d$  is the film thickness and  $T_I$  is the initial film temperature. According to the heat-transfer model, we obtained for the coefficients  $A(T_I)$  and  $B(T_I)$  at  $T_I = 60 \text{ K}$  the values  $A = 1.9 \times 10^3 \text{ K cm}^2/\text{J}$ , and  $B = 3.5 \times 10^3 \text{ K cm}^2/\text{J}$ , which change only little for higher initial film temperature  $T_I$ . For lower temperatures the strongly decreasing specific heat must be taken into account, leading to a nonlinear dependence of  $\Delta T$  on  $E_p$ , which has not been evaluated in detail. We will describe our results using an expression

$$U_N = C(\nabla T)S\nu_0 \exp[-T_p/T_F] \\ = C'E_p \exp[-T_p(BE_p + T_I)^{-1}]$$

for the Nernst signal voltage  $U_N$  with proportionality constants  $C$ ,  $C'$ , and with the relations given above for  $\nabla T$  and  $T_F$  for small  $E_p$ . For conventional type-II super-

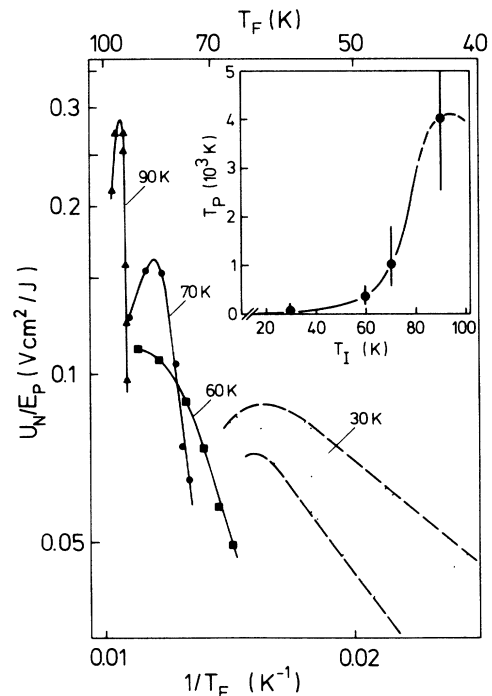


FIG. 7. Dependence of the normalized Nernst signal  $U_N/E_p$  on film temperature  $T_F$ , for various initial film temperatures  $T_I$ . The inset shows values of the pinning energy  $T_p$  obtained at different  $T_I$ .

conductors, the transport entropy  $S$  increases with temperature from zero to a maximum value at a temperature of about  $\frac{1}{2}T_c$ , depending on the material, and then decreases again to zero at  $T_c$ .<sup>16</sup> Not much change is observed in a temperature range from about  $0.3T_c$  to  $0.8T_c$ .<sup>16</sup> The detailed dependence of  $S$  on temperature for high- $T_c$  superconductors is not known, and we take  $S$  constant for the analysis of our measurements in the temperature range from 30 to 90 K.

A logarithmic plot of the Nernst voltage normalized to the laser-pulse energy  $U_N/E_P$  as a function of  $T_F^{-1}$  is shown in Fig. 7 for various temperatures  $T_I$ . At small laser-pulse energies the curves show constant slopes from which we deduce values for  $T_P$ . With increasing laser energy,  $T_F$  increases and the curves bend due to saturation of the signal. Because of uncertain  $T_F$  for the  $T_I=30$  K measurement, we give an estimated region for the slope of this curve, which is shaded in Fig. 7. The inset shows the pinning temperature  $T_P$  for the different film temperatures  $T_I$ . Our results indicate a distribution of pinning centers with different pinning energies ranging from low values,  $T_P \lesssim 100$  K, up to high values of about 4000 K. At low temperatures only loosely bound fluxons are depinned and contribute to the signal. At higher temperatures strongly bound fluxons are also thermally activated, leading to the observed strong increase of the Nernst signal.

## CONCLUSION

In conclusion, we have observed, for the first time, a Nernst effect in a high- $T_c$  superconductor by applying pulsed-laser heating of thin films. As in experiments with conventional type-II superconductors,<sup>6,16</sup> a voltage transverse to a magnetic field and transverse to a temperature gradient was observed when the sample was in the superconducting state. Because of large temperature gradients obtained by the laser-heating method, relatively large voltage signals were observed, allowing for time-resolved measurements of the thermal behavior of a high- $T_c$  film in the superconducting state.

As in conventional superconductors,<sup>17</sup> the Nernst voltage increased nearly linearly with magnetic field and showed reversed sign for reversed-field direction. With increasing sample temperature we observed a strong increase of the Nernst voltage. Assuming thermal activation of vortices, this increase in signal could not be explained by one single pinning energy. Our analysis suggests a distribution of pinning energies from 100 to 4000 K for the studied sample.

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