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Author(s)	Iimori, Toshifumi; Sabeth, Farzana; Naito, Toshio; Ohta, Nobuhiro
Citation	Journal of Physical Chemistry C, 115(48): 23998-24003
Issue Date	2011-12-08
Doc URL	http://hdl.handle.net/2115/47959
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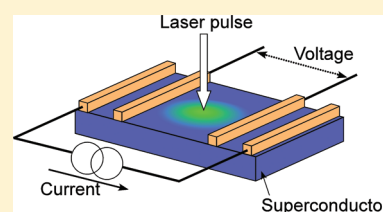
Time-Resolved Photoresponse Measurements of the Electrical Conductivity of the Quasi-Two-Dimensional Organic Superconductor β -(BEDT-TTF) $_2$ I $_3$ Using a Nanosecond Laser Pulse

Toshifumi Iimori,[†] Farzana Sabeth,[†] Toshio Naito,[‡] and Nobuhiro Ohta^{*,†}

[†]Research Institute for Electronic Science, Hokkaido University, Sapporo 001-0020, Japan

[‡]Graduate School of Science and Engineering, Ehime University, Matsuyama 790-8577, Japan

ABSTRACT: Time-resolved photoresponses in resistance have been measured following the nanosecond laser pulse excitation for the quasi-two-dimensional organic superconductors of hydrogenated and deuterated β -(BEDT-TTF) $_2$ I $_3$ [BEDT-TTF = bis(ethylenedithio)tetrathiafulvalene], which show two different superconducting states with high- T_c and low- T_c at temperatures near the critical temperatures. A transient increase of the resistance is induced by photoirradiation at all the temperatures, but a marked temperature dependence of the decay time is observed at temperatures close to the high- T_c phase transition temperature; the decay rate becomes faster and then becomes constant in both compounds, as the temperature decreases across the high- T_c phase transition temperature. The temperature dependence of the photoresponse intensity is different from the one expected from the bolometric effects, indicating the presence of the nonbolometric photoresponse. A possible mechanism explaining the photoresponse of the conductivity is discussed, based on the isotope effect on the photoresponse. A comparison is also made between β -(BEDT-TTF) $_2$ I $_3$ and κ -(BEDT-TTF) $_2$ Cu[N(CN) $_2$]Br for the transient photoresponse in resistance at temperatures across the metal-superconductor phase transition temperature.



INTRODUCTION

In modern science and technology, control of material properties by using external perturbations such as photoirradiation and an application of external electric field has been the subject of active investigation for the development of innovative optoelectronic functional materials.^{1–3} The realization of a photoinduced conversion of normal material to superconductor is, in particular, the most challenging problem. Organic conductors have attracted much attention as such potential photoswitching materials because they often show phase transitions among a variety of phases including a superconducting state upon the application of weak pressure or chemical substitution. Low electron density, electrostatic interactions of charged particles, and a transfer integral between molecules building a conduction band play important roles for the appearance of such rich electronic states.^{4–6} Recently, a photoinduced phase transition from a charge-ordered insulating phase to a metallic phase upon the irradiation of a laser pulse has been demonstrated in the organic conductor α -[bis(ethylenedithio)tetrathiafulvalene] $_2$ I $_3$ [α -(BEDT-TTF) $_2$ I $_3$] by using femtosecond pump–probe spectroscopy and time-resolved photoresponse measurements of electrical conductivity.^{7–11} However, a question still remains about the possibility of the photoinduced conversion from states of normal electrical resistivity to a superconducting state in organic conductors.

In a strategy toward realizing photoinduced superconductivity, understanding of the photoresponse of the conductivity of organic superconductors is indispensable. The quasi-two-dimensional organic superconductor β -(BEDT-TTF) $_2$ I $_3$ is an attractive

target because a slight structural change drastically influences the superconductivity. In a single crystal of β -(BEDT-TTF) $_2$ I $_3$, two superconducting states, a low- T_c state that shows a phase transition to a superconducting state at a critical temperature $T_c \cong 1.5$ K and a high- T_c state with $T_c = 7–8$ K, can coexist.^{12–14} The superconducting phase transition temperature of the crystal which initially shows $T_c \cong 1.5$ K in the low- T_c state rises to ~ 7 K at the pressure of ~ 1 kbar.¹⁴ After releasing the applied pressure (higher than 0.4 kbar) at temperatures below 125 K, a pure crystal of the high- T_c state is obtained at ambient pressure.^{15,16} Diffraction experiments have suggested that an incommensurate superlattice structure is present in the low- T_c state, and at pressures higher than 0.5 kbar, the incommensurate structure disappears.^{17,18} It is certain that the incommensurate structure is related to the disorder of a conformation of the terminal ethylene groups of the BEDT-TTF molecule.⁵

Here, we study the photoresponse of the resistance of the organic superconductor β -(BEDT-TTF) $_2$ I $_3$ at temperatures near the superconducting phase transition temperatures. In general, the relaxation from photoexcited states to the ground electronic state occurs rapidly, and almost all of the changes of material properties induced by photoexcitation become transient phenomena. Thus, the use of time-resolved measurement techniques is essential for a sensitive detection of transient changes in a nonequilibrium state after photoexcitation. In this work, we

Received: September 15, 2011

Revised: October 27, 2011

Published: October 31, 2011



report time-resolved measurement of the electrical conductivity of β -(BEDT-TTF) $_2$ I $_3$ photoexcited with a nanosecond laser pulse. The fully deuterated β -(BEDT-TTF) $_2$ I $_3$ has also been reported to show two superconducting phase transition temperatures, which are similar to those in the fully hydrogenated compound.⁵ The β -(BEDT-TTF) $_2$ I $_3$ salts are therefore suitable for the investigation of the isotope substitution effect on the photoresponse. The time-resolved measurements of photoresponse of the resistance following laser pulse excitation have also been done in fully deuterated β -(BEDT-TTF) $_2$ I $_3$. The present results of the photoresponse of resistance of β -(BEDT-TTF) $_2$ I $_3$ have also been compared with those of κ -(BEDT-TTF) $_2$ Cu[N(CN) $_2$]Br.¹⁹

EXPERIMENTAL METHODS

β -(h_8 -BEDT-TTF) $_2$ I $_3$ and β -(d_8 -BEDT-TTF) $_2$ I $_3$ were studied in this work, where h_8 -BEDT-TTF and d_8 -BEDT-TTF indicate fully hydrogenated and deuterated BEDT-TTF molecules, respectively. Single crystals were prepared by using a galvanostatic electrocrystallization method with BEDT-TTF, tetrabutylammonium triiodide, and 1,1,2-trichloroethane as a solvent. h_8 -BEDT-TTF and d_8 -BEDT-TTF (Tokyo Chemical Industry) were used as received. The dimension of a typical sample used in the present study was $1 \times 0.4 \times 0.1$ mm³. The time-resolved measurement system of the electrical conductivity in organic superconductors has been reported elsewhere.¹⁹ Briefly, the sample was placed in a cryostat, which can control temperature in the range between 1.5 and 300 K using temperature-controlled helium buffer gas. Gold wire (25 or 10 μ m in diameter) and gold paste were used as electrodes on the conducting a - b plane of the crystal. The resistance was measured using the four-wire connection technique. In a steady-state measurement of the temperature dependence of the resistance, the current-reversal method was used with a combination of a current source (Keithley, model 2400) and a nanovolt meter (Keithley, model 2182). The cooling rate at 100 K was 0.38 K min⁻¹. Hereafter, the resistance at temperature T is denoted by $R(T)$. In time-resolved measurements of the resistance, a constant current of 1 mA with a time-width of 80 ms was used, and the attending voltage of the sample was amplified and detected by a digital oscilloscope (LeCroy, 1 GHz bandwidth). The transient change in the resistance was obtained from the voltage waveform divided by the magnitude of the bias current.

As a light source, we employed an optical parametric oscillator unit mounting a BBO crystal pumped by a third harmonic of the output of a pulsed Nd:YAG laser (QuantaRay, LAB-150). The pulse width of the output was ~ 10 ns, and the wavelength of the irradiation light was 532 nm. An optical fiber was used to deliver the laser pulse to the center of the crystal surface between the two electrodes for the voltage measurement. The diameter of the illuminated area was ~ 0.4 mm. The direction of the propagation of the laser light was normal to the conducting plane of the crystal. The repetition rate of the laser pulse was 2 Hz, but a single shot of the laser pulse was used to obtain the signal.

When the laser pulse was irradiated to the sample, a sharp voltage signal was observed in synchronization with the photoirradiation even without the bias current. For the measurements of the signal due to the photoinduced change of resistance, such a photovoltaic component was subtracted from the observed voltage waveform. Moreover, the dark resistance measured by using the pulsed bias current was identical to that obtained by the

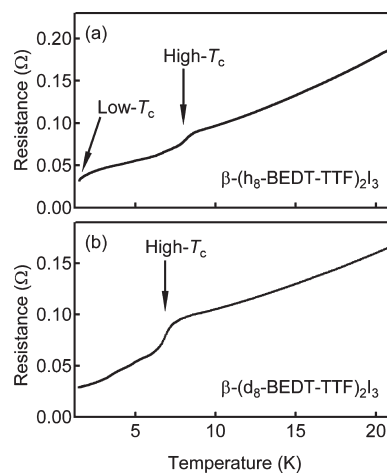


Figure 1. (a) Resistance of β -(h_8 -BEDT-TTF) $_2$ I $_3$ as a function of temperature. The arrows indicate two different phase transition temperatures. (b) Resistance of β -(d_8 -BEDT-TTF) $_2$ I $_3$ as a function of temperature.

standard steady-state measurement technique with a smaller current. This observation indicates that the effects of the heating due to the bias current and thermoelectric voltages, which cause an error in the voltage measurements, are negligible in the time-resolved experiments.

RESULTS AND DISCUSSION

The coexistence of the two superconducting phase transitions arising from the high- T_c and low- T_c states has been reported both in β -(h_8 -BEDT-TTF) $_2$ I $_3$ and in β -(d_8 -BEDT-TTF) $_2$ I $_3$, as described in the introduction. The two superconducting transitions of the former crystals are exemplified in the plot of resistance as a function of temperature in Figure 1a. A step appearing near 7.5 K indicates the presence of the high- T_c state. In addition, the drop of the resistance begins at temperatures near 1.5 K, which is ascribed to the superconducting phase transition to the low- T_c state.

Time profiles of the change in resistance ($\Delta R(t)$) following photoirradiation have been measured at various temperatures across the high- T_c phase transition temperature. The results are shown in Figure 2. The horizontal axis represents the time elapsed after the irradiation of the laser pulse at time $t = 0$. The vertical axis represents the change from the original resistance without photoirradiation, and thus, the signal in the positive direction indicates the photoinduced increase of the resistance. Although the resistance is increased by photoirradiation at all the temperatures, the shape of the time profile depends on temperature. Actually, the time profiles show multiexponential decays, implying the presence of plural states having a nonequilibrium condition. In the present analysis, however, the decay time (τ) was calculated by assuming a single exponential decay; the value of τ was obtained by the integral of $\Delta R(t)$ divided by the peak intensity (ΔR_{peak}) of $\Delta R(t)$ with the following equation:

$$\tau = \frac{1}{\Delta R_{\text{peak}}} \int_0^{\infty} \Delta R(t) dt \quad (1)$$

The value of τ can be a measure of the decay time of the photoinduced change in the resistance. Plots of τ as a function of temperature are shown in Figure 3. Although there was a variation

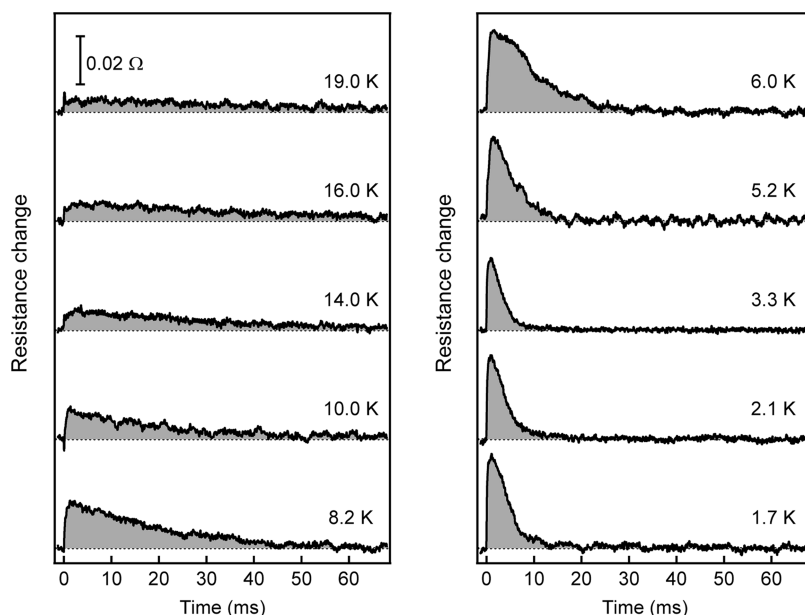


Figure 2. Time profiles of the photoinduced change of resistance in β -(h_8 -BEDT-TTF) $_2$ I $_3$. The vertical bar on the top panel is a scale of the resistance change. The irradiated laser light intensity was 3×10^{-6} J/pulse.

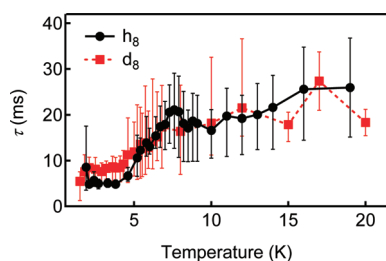


Figure 3. Plot of decay times (τ) of the photoinduced change of resistance versus temperature. The labels h_8 and d_8 represent the results for β -(h_8 -BEDT-TTF) $_2$ I $_3$ and β -(d_8 -BEDT-TTF) $_2$ I $_3$, respectively.

between different specimens, a small value of τ on the order of a few milliseconds was obtained in the high- T_c state at temperatures below 5 K. At temperatures above 5 K, τ increases with the increase of temperature toward the high- T_c phase transition temperature. Amplitudes of the $\Delta R(t)$ also show temperature dependence (Figure 2). At temperatures above the high- T_c , i.e., in the metallic phase, the amplitudes were smaller than those observed in the superconducting phase.

The resistance of β -(d_8 -BEDT-TTF) $_2$ I $_3$ as a function of temperature is shown in Figure 1b. The phase transition to the high- T_c superconducting phase was observed at 6.5–7 K. The magnetic susceptibility study has suggested a normal shift (decrease) of the high- T_c phase transition temperature upon deuteration of the BEDT-TTF molecule, although the observed shift contains considerable uncertainty.²⁰ In fact, a slight decrease of the high- T_c temperature was observed with deuteration (see Figure 1b), consistent with the previous result. The time profiles of $\Delta R(t)$ are shown in Figure 4. The decay time obtained with eq 1 is shown in Figure 3. The temperature dependence of the decay time across the high- T_c temperature is analogous to that observed in β -(h_8 -BEDT-TTF) $_2$ I $_3$.

Temperature Dependence of Decay Times and Mechanism of Photoresponse. A significant temperature dependence

of τ , i.e., the photoresponse decay times of the resistance, was observed in the range from 4 to 8 K (see Figure 3), although τ shows minor and subtle temperature dependence both in the metallic phase at temperatures above 8 K and in the superconducting phase at temperatures below 4 K. As the temperature falls below the high- T_c , the decay time shows a steep decrease. It is unlikely that the temperature rise of the crystal induced by the absorption of the pulsed laser light could explain such a marked change in the vicinity of T_c . One should consider the system at nonequilibrium or nonbolometric effects as the origin of the photoresponse.

Mechanisms of nonbolometric photoresponse have been discussed extensively for high- T_c superconducting materials.^{21,22} In granular or polycrystalline superconducting thin films, the origin of nonbolometric photoresponses is thought to be the photoinduced modulation of current flowing through a weakly coupled grain boundary.^{23,24} In the present experiment, however, such a mechanism is unlikely because we used single crystals in which grain boundaries were nonexistent.

In conventional superconductors, a breakup of Cooper pairs induced by photoexcitation results in a nonequilibrium superconductivity.²⁵ Indeed, a change of the superconducting gap after a laser pulse excitation has been demonstrated experimentally.^{26,27} Ultrafast dynamics of electrons in high- T_c copper oxides was also investigated with several different methods.²⁸ Pump–probe spectroscopic studies of YBa $_2$ Cu $_3$ O $_{7-\delta}$ single crystals using femtosecond laser pulse excitation at temperatures below their T_c have shown that the relaxation of the quasiparticles generated after photoexcitation occurs on a picosecond time scale.^{29–31}

In the recombination process of the quasiparticles, excess energy is released as phonons with the energy greater than 2Δ , which is the superconducting energy gap. These phonons can, however, break Cooper pairs, and consequently, the decay process of the phonons is a rate-determining step or a bottleneck step.^{28,32} Considering the phonon scattering due to the

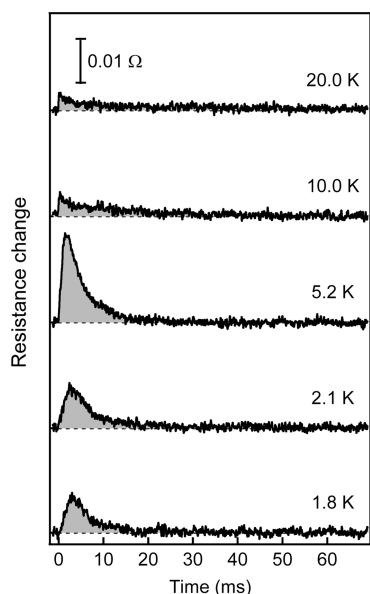


Figure 4. Time profiles of the photoinduced change of resistance in β -(d_8 -BEDT-TTF) $_2$ I $_3$. The vertical bar on the top panel is a scale of the resistance change. The irradiated laser light intensity was 9×10^{-6} J/pulse.

anharmonic coupling between phonons, Kabanov et al. have derived the expression describing the decay rate of the phonon in the following form:

$$\frac{1}{\tau_{\text{ph}}} = \frac{9\pi\nu^2 w^2 k_B T_q \Delta(T_L)}{(\hbar\Omega_c)^3} \quad (2)$$

where τ_{ph} is the decay time of the phonon, ν is the effective number of phonon modes per unit cell participating in the relaxation, w is the anharmonic coupling constant between phonon modes, k_B is Boltzmann's constant, T_q is the temperature of quasiparticles and phonons with the energy greater than 2Δ , $\Delta(T_L)$ is the gap at the lattice temperature, T_L , \hbar is Planck's constant divided by 2π , and Ω_c is the cutoff frequency in the Debye approximation.³³ Note that the contribution of the Ω_c term originates from the phonon density of states. Theoretical investigation of low frequency phonons in β -(BEDT-TTF) $_2$ I $_3$ has suggested a considerable contribution of intermolecular vibrational modes such as librational and translational motions of BEDT-TTF molecules.³⁴ In addition, extensive mixing between these intermolecular vibrational modes and intramolecular vibrational modes has also been indicated. One can incontrovertibly expect the isotope effect on the frequencies of these vibrational modes and the relevant phonon density of states. Hence, it is likely that the decay time due to the recombination process shows the remarkable isotope effect. In the present study of β -(BEDT-TTF) $_2$ I $_3$, however, the isotope effect was ambiguous (Figure 3), and the observed dynamics was in a millisecond time scale, which is much slower in comparison with the previous results for other superconductors. In conclusion, we could not obtain the positive proof in the deuterium effect to argue that the photoresponse of the resistance is ascribed to the recombination process of the quasiparticles.

In superconducting materials, enhancement of the flux creep and flux motion is another source of resistance increase.^{21,35,36}

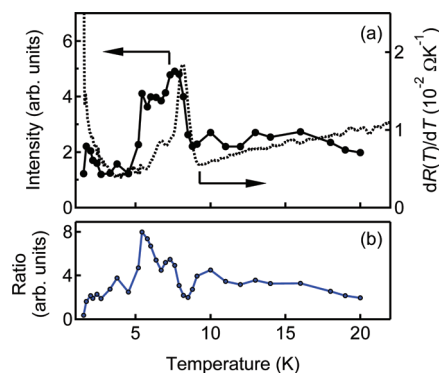


Figure 5. (a) Integrated intensity of the time profile of the resistance change as a function of temperature (solid line) and a derivative of the resistance-versus-temperature curve $[R(T)]$ (dotted line) of β -(h_8 -BEDT-TTF) $_2$ I $_3$ shown in Figure 1. (b) Integrated intensity divided by the derivative of $R(T)$ as a function of temperature.

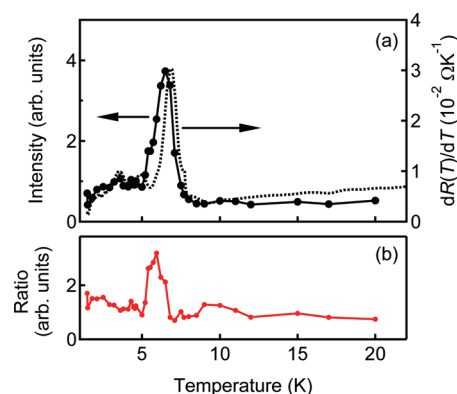


Figure 6. (a) Integrated intensity of the time profile of the resistance change as a function of temperature (solid line) and a derivative of the resistance-versus-temperature curve $[R(T)]$ (dotted line) of β -(d_8 -BEDT-TTF) $_2$ I $_3$ shown in Figure 1. (b) Integrated intensity divided by the derivative of $R(T)$ as a function of temperature.

Thus, photoactivation of flux creep and flux motion may be considered as the origin of the observed photoinduced increase in resistance. Other explanations considering the photoinduced creation of a pair of vortices³⁷ or the change of the impedance of superconducting materials caused by the modulation of the Cooper pair density³⁸ may also have to be considered.

Temperature Dependence of Photoresponse Intensity. As mentioned above, it is likely that the photoresponse is ascribed to the nonbolometric effect. We have further examined the possibility of the bolometric effect from the point of view of the photoresponse intensity.

The bolometric photoresponse of the resistance caused by a temperature increase (ΔT) is given by

$$\Delta R_s = \frac{dR(T)}{dT} \Delta T \quad (3)$$

where ΔR_s is the signal intensity, depending both on the derivative of $R(T)$ and on ΔT . In Figure 5a, we show the integrated intensity of the observed time profile $\Delta R(t)$ of β -(h_8 -BEDT-TTF) $_2$ I $_3$ as a function of temperature. The derivative of $R(T)$ in Figure 1 is also shown in Figure 5a. In this analysis, we have used the integrated intensity as the signal intensity, not

the value of ΔR_{peak} , i.e., $\Delta R_s = \int_0^\infty \Delta R(t) dt$, because the integrated intensity is linearly proportional to the light intensity. As the temperature decreases from 20 to 9 K in the normal metallic state, the integrated intensity is nearly constant. As the temperature further decreases, the crystal shows the phase transition to the high- T_c state, and the integrated intensity of $\Delta R(t)$ increases with a maximum near 7–8 K (see solid line in Figure 5a). Below these temperatures, the integrated intensity decreases to a level close to that of the metallic state. The shapes of both the integrated intensity and the derivative of $R(T)$ are different from each other in the region from 5 to 7 K, as clearly recognized in Figure 5b, which shows a peak in this temperature range.

The results for β -(d_8 -BEDT-TTF) $_2$ I $_3$ (Figure 6a,b) similarly show an enhancement of the value of the ratio at temperatures below the high- T_c phase transition temperature. Thus, one can find the similarity of the temperature dependence of the ratio near the high- T_c phase transition temperature, irrespective of the isotopic composition of the specimen.

If pulsed laser light induces the crystal temperature to rise, the corresponding increase can be calculated from the following thermodynamic relationship:

$$\Delta E = \int_{T_1}^{T_2} C(T) dT \quad (4)$$

where ΔE is the energy of the laser pulse, T_1 and T_2 are the temperatures of the crystal before and after the photoirradiation, respectively, and $C(T)$ is the heat capacity at T . The specific heat as a function of temperature shows a smooth variation over the temperatures measured in this experiment, and it monotonically decreases with the lowering of temperature.^{39,40} This result then leads to the monotonic increase of $\Delta T = T_2 - T_1$ by photoirradiation with the lowering temperature. According to eq 3, the ratio of ΔR_s relative to the derivative of $R(T)$ is proportional to ΔT , if the observed photoresponse results from the bolometric effect. Actually, the observed ratios (Figures 5b and 6b) do not show such a monotonic change as a function of temperature. This result confirms that the bolometric effect is insignificant in the photoresponse.

Comparison of the Present Results with Those of the Organic Superconductor κ -(BEDT-TTF) $_2$ Cu[N(CN) $_2$]Br. A time-resolved measurement of the photoinduced change of the electrical conductivity in the organic superconductor κ -(BEDT-TTF) $_2$ Cu[N(CN) $_2$]Br (κ -Br) has been reported by using a technique similar to the present study.¹⁹ The critical temperature of κ -Br is ~ 12 K, and the resistivity increases with the irradiation of a nanosecond laser pulse at temperatures near this T_c . A nonbolometric component exists in the photoresponse of the superconducting state. Decay times of the photoinduced resistance change are remarkably shorter in the normal metallic state, exhibiting an anomalous increase at the temperature of ~ 10 K below the T_c .

The presence of the nonbolometric component in the superconducting state seems to be common in the photoresponse of the two organic superconductors of β -(BEDT-TTF) $_2$ I $_3$ and κ -Br. However, the decay time hardly shows an anomalous increase at temperatures near the high- T_c transition temperature of β -(BEDT-TTF) $_2$ I $_3$ (Figure 3), which is in strict contrast with κ -Br. It is probable that the weak anomaly of β -(BEDT-TTF) $_2$ I $_3$ is partly related to its small percentage of the superconducting volume, though the evaluation of the volume percentage of the high- T_c superconducting state of β -(BEDT-TTF) $_2$ I $_3$ from the

resistivity as a function of temperature is not straightforward. Studies of the magnetic susceptibility have shown that β -(BEDT-TTF) $_2$ I $_3$ shows a diamagnetic signal of $\sim 60\%$ and at least 25% of that of a perfect superconductor at $T = 0.115$ K and $T < 0.8$ K, respectively.^{39,41} It is likely that the percentage of the superconducting volume at the temperatures used in this work is much smaller than these values. However, the percentage of the superconducting volume of κ -Br is close to 100% at $T < 10$ K irrespective of the cooling rate.⁴² Consequently, the appearance of the nonbolometric component of the photoresponse intensity might be sensitive to the presence of the superconducting state, and the relaxation dynamics of the photoresponse might have a weaker sensitivity to the percentage of the superconducting volume.

SUMMARY

The photoinduced change of the electrical conductivity of β -(h_8 -BEDT-TTF) $_2$ I $_3$ and β -(d_8 -BEDT-TTF) $_2$ I $_3$ has been studied by using a time-resolved measurement technique with photoirradiation of a nanosecond laser pulse. These salts show two different superconducting phase transitions at ambient pressure. Time profiles of the photoinduced resistance change have been measured at temperatures near the two superconducting phase transition temperatures, and the transient increase of the resistance is induced by photoirradiation at all the temperatures under the present study. The decay times of the resistance change are longer in the normal metallic phase than those at temperatures below the high- T_c transition temperature. The marked temperature dependence of the decay time suggests nonbolometric effects as the origin of the photoresponse. On the basis of the isotope effect, a possible mechanism to explain the observed photoresponse is argued. The magnitude of the ratio between the photoresponse signal intensity and the derivative of $R(T)$ versus T increases at temperatures lower than the high- T_c superconducting transition temperature. This result allows us to confirm the contribution of a nonbolometric component in the observed photoresponse. It has also been discussed that the temperature dependence of the photoresponse decay time of β -(BEDT-TTF) $_2$ I $_3$ is different from that of κ -Br.

AUTHOR INFORMATION

Corresponding Author

*E-mail: nohta@es.hokudai.ac.jp.

ACKNOWLEDGMENT

This work has been supported by a Grant-in-Aid for Scientific Research (A) (Grant No. 20043005) from the Ministry of Education, Culture, Sports, Science, and Technology in Japan.

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