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Carrier relaxation dynamics in the organic superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ under pressure

Satoshi Tsuchiya · Koichi Nakagawa · Jun-ichi Yamada · Yasunori Toda

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Abstract Photo-induced carrier relaxation dynamics have been investigated for the organic superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ with different probe polarization under 1.3 kbar at low temperatures. We successfully observed the isotropic and anisotropic responses for the probe polarization, which were found to appear at 56 K. By comparing the responses with and without applying pressure, we found that those were slightly changed, indicating that the application of pressure has an effect on the carrier relaxation dynamics.

Keywords organic superconductor · time-resolved spectroscopy · high-pressure

PACS 78.47.D- · 74.70.Kn

1 Introduction

Organic molecular materials have attracted much interest from the point of view of fundamental physics in a strongly correlated electron system. A rich variety of electronic phases such as superconductivity (SC) and charge/spin-density-waves (CDW/SDW) are induced by changes of temperature and pressure[**?**,**?**,**?**]. Since the organic materials are relatively softer than transition metal oxides, structural modifications and/or electronic phase transitions appear under low or moderate pressures. For example, κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl shows the insulator-metal transition with applying pressure only above 300 bar and superconductivity below T_c = 12.8 K[**?**,**?**]. Moreover, the phase diagram of κ-(BEDT-TTF)$_2$X family, where X corresponds to anion molecules, is quite similar to that of the cuprate superconductors if pressure is replaced by carrier doping[**?**,**?**]. Since application of pressure corresponds to change of correlation between conducting electrons, the pressure-induced study in the organic materials will provide another aspect to the underlying physics in the cuprates in terms of strong electron correlation, leading to full understanding of mechanism of high-temperature superconductivity.

Femtosecond time-resolved spectroscopy is a powerful technique to measure relaxation of photo-excited quasi-particles (QPs) resolved in the time domain. This measurement has made a significant contribution to researches of the coexistence and competition of SC, CDW/SDW and pseudogap[**?**,**?**,**?**,**?**,**?**,**?**,**?**,**?**,**?**,**?**,**?**,**?**,**?**] since those relaxation dynamics can be observed individually by differences of relaxation time, probe-polarization, the wavelength and intensity of light and so on. However, for the organic compounds, systematic measurements have been limited[**?**,**?**,**?**,**?**] and not been performed with applying pressure because of the difficulties of handling for the pressure-induced equipment for the optical measurements.

Recently, we have successfully developed a pump-probe measurement system under high-pressure and low temperature with a piston-cylinder type pressure cell and an optical fiber bundle[**?**]. This pressure cell can allow us to perform the systematic study easily with varying pressure for organic superconductors as compared to a diamond anvil cell. In this paper, we report the carrier relaxation dynamics in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ (κ-NCS) under pressure resolved by different probe-polarizations. Anisotropic response for the probe polar-

S. Tsuchiya · K. Nakagawa · Y. Toda
Depertment of Applied Physics, Hokkaido University, Hokkaido 060-8628, Japan
Tel.: +81-11-706-6630
Fax: +81-11-706-6630
E-mail: satoshi.tsuchiya@eng.hokudai.ac.jp

J. Yamada
Graduate School of Material Science, University of Hyogo, Hyogo 650-004, Japan

ization, which was similar to that obtained in the measurement without applying pressure, has been observed even under the pressure of 1.3 kbar. By decomposing the data into anisotropic and isotropic components, we evaluated the decay time of the relaxation in the each component and compared with the result under ambient pressure.

2 Experimental

Single crystals of κ-NCS was prepared electrochemically[**?**]. The crystal consists of alternate stacking of the conducting layer of BEDT-TTF molecules and insulating layer of $Cu(NCS)_2$. In the κ-$(BEDT\text{-}TTF)_2X$ family, the electronic state is controlled by pressure or chemical substitution of anion molecules X[**?**]. It has been pointed out that the control of these parameter modulates effective electron correlation t/U, where t and U mean the transfer integral between dimers of BEDT-TTF molecules and the on-site Coulomb repulsion, respectively. The κ-NCS salt becomes a superconductor below $T_c \sim 10$ K under ambient pressure. Application of pressure increases t/U, leading to diminishment of the electron correlation effects. The organic sample whose size in the present experiments was 500 $\times$ 200 $\times$ 50 μm^3, was set on the end surface of the optical fiber bundle and installed in a piston-cylinder type pressure cell. The pressure was calibrated by the Ruby fluorescence method at room temperature. The detail of experimental setup is referred to the previous report[**?**].

The optical measurements were performed using 120 fs pulses centered at 400 nm for a pump (~ 1.3 mJ/cm^2) and 800 nm for a probe from a cavity-dumped Ti:sapphire oscillator with a repetition rate of 54 kHz to avoid the heating effect. The pump and probe beams were coaxially overlapped by a dichroic mirror and irradiated perpendicular to the conducting plane of the sample in the pressure cell through the fiber bundle. In this system, broadening and change of polarization of pulse owing to the dispersions in the fiber bundle are negligible[**?**]. A polarization dependence of the probe is measured by rotating a half-wave plate, where θ is measured from the b axis. Since optical penetration depth is roughly estimated as 13 μm for pump and 3 μm for probe in κ-NCS, respectively[**?**], bulk electronic properties are measurable.

3 Results and Discussion

Figures **??**(a) and (b) show a transient reflectivity change $\Delta R/R$ for $\theta = 0^\circ$ and intensity plot as a function of θ in κ-NCS at 56 K under 1.3 kbar. The polar plot of the maximum values of $\Delta R/R$ is shown in Fig. **??** (c). $\Delta R/R$ shows anisotropic whose signal enhanced along the direction 0° and 180°, which is similar to that at 43 K in the previous report[**?**]. The emergence of the anisotropic response can be attributed to spatial symmetry breaking due to the glass-like structural transition at 70 K[**?**].

To analyze the data further, we decomposed the data into the anisotropic and isotropic components by fitting the following equation

$$\frac{\Delta R(\theta)}{R} \propto \Delta R_{\rm ani} \cos(2(\theta - \phi_{\rm c})) + \Delta R_{\rm iso}, \qquad (1)$$

to the angular dependence of $\Delta R(\theta)/R$, where $\Delta R_{\rm ani}$, $\Delta R_{\rm iso}$ and $\phi_{\rm c}$ denote anisotropic and isotropic components of $\Delta R/R$ and an initial phase, respectively. The initial phase is assumed as constant. Figures **??**(a) and (b) present the reflectivity changes normalized by the maximum value of $\Delta R/R$ for the isotropic and anisotropic components, respectively, for various temperatures. The data at 43 K are obtained from the previous report[**?**].

To explore pressure effect on carrier relaxation dynamics, we present the isotropic and anisotropic transient responses which were obtained under ambient pressure at 42 and 57 K in Figs. **??**(c) and (d), respectively, for comparison and investigated decay time. Figure **??** shows the temperature dependences of the decay time obtained by fitting the rapid decline of the transient with a single-exponential function for 0 and 1.3 kbar. Under 0 kbar at 42 K, the fitting results yielded $\tau \sim$ 0.79 and 1.06 ps for the isotropic and anisotropic components, respectively. On the other hand, τ were estimated as 1.11 and 0.63 ps under 1.3 kbar at 43 K, indicating that τ were changed in the opposite direction for pressure. The similar trend is also observed at around 56 K. The τ changes from 0.85 to 1.30 ps in the isotropic component but from 0.92 to 0.88 ps in the anisotropic component with increasing pressure.

Let us discuss origin of the variation of τ. In our results, τ of the isotropic component reduced, while, oppositely, that of the anisotropic component became large by application of pressure. The behaviors suggest that pressure affects each the isotropic and anisotropic QP relaxation dynamics in a different way. Generally, pressure modulates the electron-phonon (*e-ph*) interaction. Since the carrier relaxation rate $1/\tau$ is proportional to the strength of the *e-ph* interaction[**?**,**?**], the applied pressure may lead to reduction of the decay

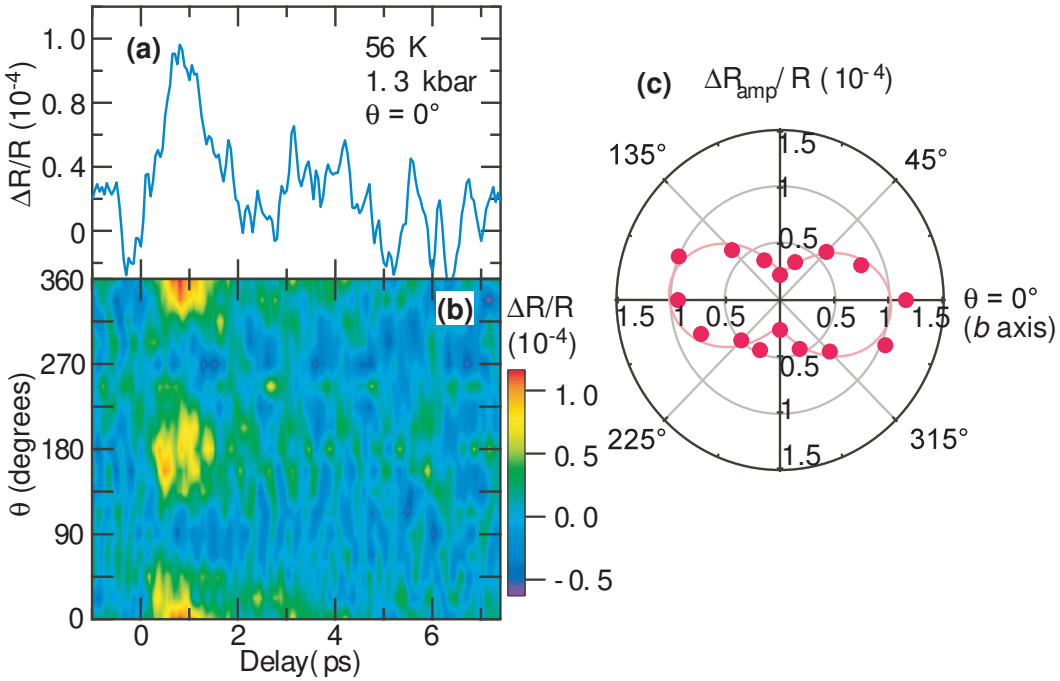


Fig. 1 (a), (b)Transient change of reflectivity $\Delta R/R$ for $\theta = 0°$ and intensity plots of $\Delta R/R$ as a function of probe-polarization-angle under 1.3 kbar at 56 K for $F \sim 1.3$ mJ/cm^2, respectively. (c) Polar plots of the maximum values of $\Delta R/R$ under 1.3 kbar at 56 K. The solid lines indicate the results fitted by a double-angle function.

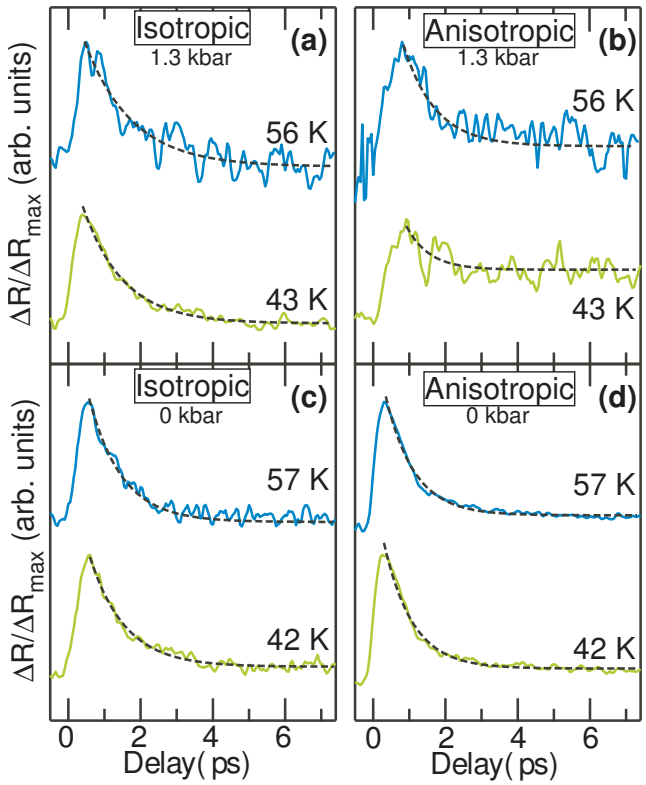


Fig. 2 (a), (b) and (c), (d) Normalized $\Delta R/R$ transients of isotropic and anisotropic components for various temperatures under 1.3 kbar and 0 kbar, respectively. The data are shifted for clarity.

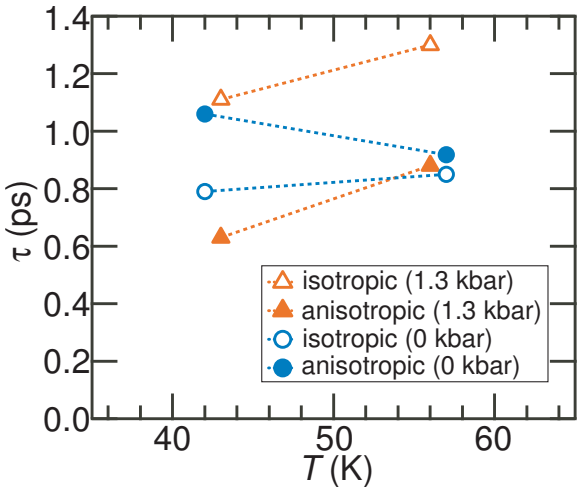


Fig. 3 Temperature dependence of the decay time under 0 and 1.3 kbar.

time of the QP relaxation. Thus, this can explain the change of τ for the anisotropic response. Indeed, such lattice effect can be crucial for a momentum dependent anisotropy in terms of QPs dynamics in the cuprates[**?**,**?**]. For the isotropic response, modulation of other parameter, such as the effective electron correlation t/U caused by application of pressure may have a crucial influence on the QP relaxation dynamics.

4 Summary

By performing polarized femtosecond pump-probe spectroscopy on κ-NCS in the piston-cylinder type pressure cell, we have successfully observed anisotropic carrier relaxation dynamics at 1.3 kbar at 56 K. From the probe polarization dependence, the decay times of the relaxation for the isotropic and anisotropic components were estimated and compared with those obtained under ambient pressure. As pressure increased, the decay time increased for the isotropic component, while decreased for the anisotropic component. The results suggest that application of pressure has effect on each the components of the carrier relaxation dynamics in the opposite way.

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References

1. D. Jerome, A. Mazaud, M. Ribault, and K. Bechgaard, J. Phys., Lett. **41**, L95 (1980).
2. P. Limelette, P. Wzietek, S. Florens, A. Georges, T. Costi, C. Pasquier, D. Jerome, C. Meziere, and P. Batail, Phys. Rev. Lett. **91**, 016401 (2003).
3. B. J. Powell and R. H. McKenzie, J. Phys.: Condens. Matter **18**, R827 (2006).
4. R. H. McKenzie, Science **278**, 820 (1997)
5. A. Rothwarf and B. N. Taylor, Phys. Rev. Lett. **19**, 27 (1967).
6. V. V. Kabanov, J. Demsar, B. Podobnik, and D. Mihailovic, Phys. Rev. B **59**, 1497 (1999).
7. J. Demsar, B. Podobnik, V. V. Kabanov, Th. Wolf, and D. Mihailovic, Phys. Rev. Lett. **82**, 4918 (1999).
8. R. A. Kaindl, M. Woerner, T. Elsaesser, D. Smith, J. Ryan, G. Farnan, M. McCurry, and D. Walmsley, Science **287**, 470 (2000).
9. R. D. Averitt, G. Rodriguez, A. I. Lobad, J. L. W. Siders, S. A. Trugman, and A. J. Taylor, Phys. Rev. B **63**, 140502(R) (2001).
10. D. Dvorsek, V. V. Kabanov, J. Demsar, S. M. Kazakov, J. Karpinski, and D. Mihailovic, Phys. Rev. B **66**, 020510 (2002).
11. P. Kusar, V. V. Kabanov, S. Sugai, J. Demsar, T. Mertelj, and D. Mihailovic, Phys. Rev. Lett. **101**, 227001 (2008).
12. T. Mertelj, V. V. Kabanov, C. Gadermaier, N. D. Zhigadlo, S. Katrych, J. Karpinski, and D. Mihailovic, Phys. Rev. Lett. **102**, 117002 (2009).
13. C. Gadermaier, A. S. Alexandrov, V. V. Kabanov, P. Kusar, T. Mertelj, X. Yao, C. Manzoni, D. Brida, G. Cerullo, and D. Mihailovic, Phys. Rev. Lett. **105**, 257001 (2010).
14. C. Giannetti, F. Cilento, S. Dal Conte, G. Coslovich, G. Ferrini, H. Molegraaf, M. Raichle, R. Liang, H. Eisaki, M. Greven, A. Damascelli, D. van der Marel, and F. Parmigiani, Nat. Commun. **2**, 353 (2011).
15. Y. Toda, F. Kawanokami, T. Kurosawa, M. Oda, I. Madan, T. Mertelj, V. V. Kabanov, D. Mihailovic, Phys. Rev. B **90**, 094513 (2014).
16. S. Tsuchiya, Y. Sugawara, S. Tanda and Y. Toda, J. Opt. **17**, 085501 (2015).
17. T. Mertelj, P. Kusar, V. V. Kabanov, L. Stojchevska, N. D. Zhigadlo, S. Katrych, Z. Bukowski, J. Karpinski, S. Weyeneth, and D. Mihailovic, Phys. Rev. B **81**, 224504 (2010).
18. T. Naito, Y. Yamada, T. Inabe, and Y. Toda, J. Phys. Soc. Jpn. **77**, 064709 (2008).
19. T. Naito, M. Fukusaku, S. Matsuo, K. Tajima, T. Inabe, Y. Toda, Physica B **405**, S269 (2010).
20. Y. Toda, T. Mertelj, T. Naito, and D. Mihailovic, Phys. Rev. Lett. **107**, 227002 (2011).
21. T. Naito, S. Matsuo, T. Inabe, Y. Toda, J. Supercond. Nov. Magn. **26**, 1679 (2013).
22. S. Tsuchiya, Y. Kino, K. Nakagawa, D. Nakagawa J. Yamada and Y. Toda, Rev. Sci. Instrum. 87, 043104 (2016).
23. H. Anzai, J. M. Delrieu, S. Takasaki, S. Nakatsuji a and J. Yamada, J. Cryst. Growth **154**, 145 (1995).
24. A. Ugawa G. Ojima, K. Yakushi, and H. Kuroda, Phys. Rev. B **38**, 5122 (1988)
25. J. Muller M. Lang F. Steglich J. A. Schlueter, A. M. Kini, and T. Sasaki Phys. Rev. B **81**, 224504 (2010).
26. R. H. M. Groeneveld, R. Sprik, and A. Lagendijk, Phys. Rev. B **51**, 11433 (1995).
27. V. V. Kabanov and A. S. Alexandrov, Phys. Rev. B **78**, 174514 (2008).
28. J. Graf, C. Jozwiak, C. L. Smallwood, H. Eisaki, R. A. Kaindl, D. H. Lee, and A. Lanzara, Nature Physics **7**, 805 (2011).
29. C. L. Smallwood, J. P. Hinton, C. Jozwiak, W. Zhang, J. D. Koralek, H. Eisaki, D. H. Lee, J. Orenstein, and A. Lanzara, Science **336**, 1137 (2012).