

# Low-Temperature Specific Heat in the Charge-Density-Wave State of $\text{ZrTe}_3$ and $\text{NbTe}_4$

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## Abstract

Low-temperature specific heat of CDW compounds  $\text{ZrTe}_3$  and  $\text{NbTe}_4$ , measured between 1.5 K and 10 K, exhibited an anomaly which deviates from the regular phonon term; an excess specific-heat contribution appeared around  $T=6$  K. The excess contribution was well presented in terms of the phason contribution of CDW. The pinning frequency estimated from the analysis was quite high in comparison to the value of common CDW conductors, which is not inconsistent with the linear relation between current and voltage.

*Key words:* low dimensional conductor; CDW; phason excitations; specific heat

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## 1. Introduction

In many charge-density-wave (CDW) compounds such as  $(\text{TaSe}_4)_2\text{I}$ , KCP, and blue bronze, a specific heat ( $C$ ) anomaly which deviates from the regular phonon term with a  $T^3$  law has been observed at low temperatures [1-3]. The anomaly is an excess specific heat with a bump in the relation between  $C/T^3$  and  $T$ , which is generally interpreted in terms of a contribution of CDW phase excitations or phason modes. The contribution due to the phason modes is well described by a modified Debye excitation spectrum with two cutoff frequencies of the pinning frequency for the pinned CDW and the "phason Debye frequency" [1, 2]. Thus, we can investigate properties of the pinned CDW through the contribution of phasons to the low-temperature specific heat. In fact, it is reported that the value of the pinning frequency estimated from the specific heat is consistent with that obtained from a measurement of nonlinear conduction in the pinned CDW [1-3]. Here we report the broad bump in the

$C/T^3$  observed in the low-temperature specific heat of CDW compounds  $\text{ZrTe}_3$  and  $\text{NbTe}_4$  [4, 5]. The broad bump is found to be well described in terms of the phason contribution of CDW.

## 2. Experiments

The  $\text{ZrTe}_3$  and  $\text{NbTe}_4$  single crystals were grown by vapor transport in sealed quartz tube using iodine as transport agent. The specific heat was measured by the thermal relaxation method in the temperature range from 1.5 to about 10 K using a silicon-on-sapphire bolometer. The quantity of the crystals used in the specific-heat measurement was about 5 mg.

## 3. Results and discussion

Figure 1 shows the temperature dependence of the specific heat ( $C$ ) of  $\text{ZrTe}_3$  divided by  $T^3$ . With increasing temperature,  $C/T^3$  decreases wholly with a broad

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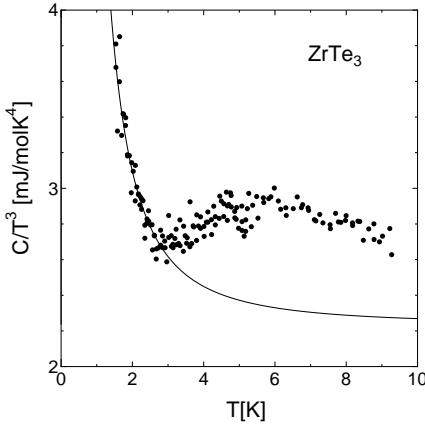


Fig. 1. Temperature dependence of the specific heat of ZrTe<sub>3</sub> in a  $C/T^3$  vs.  $T$  plot. Solid line is described by the function of  $C/T^3 = \gamma/T^2 + \beta$ .

bump around 6 K. The specific heat below 3 K can be well presented by two terms of the electronic specific heat ( $\gamma T$ ) and the Debye specific heat ( $\beta T^3$ ). Solid line in Fig. 1 is described by the function of  $C/T^3 = \gamma/T^2 + \beta$ . The values of  $\gamma$  and  $\beta$  obtained agree well with previous result [6]. The solid line deviates from experimental points above 3 K. An excess contribution,  $C_{\text{ex}}/T^3$  ( $= C/T^3 - (\gamma/T^2 + \beta)$ ) is seen around 6 K.

Figure 2 shows the temperature dependence of the specific heat of NbTe<sub>4</sub> divided by  $T^3$ . With increasing temperature,  $C/T^3$  decreases initially, but above 3.5 K it turns to increase with a smooth bump around 6 K. The increase in  $C/T^3$  observed in the high-temperature range above 7 K indicates that the  $C$  of NbTe<sub>4</sub> cannot be simply represented by only two terms of  $\gamma T$  and  $\beta T^3$ . We assume the contribution from anharmonic lattice vibration ( $\sigma T^5$ ). Solid line in Fig. 2 is described by the function of  $C/T^3 = \gamma/T^2 + \beta + \sigma T^2$ . We find an excess contribution,  $C_{\text{ex}}/T^3$  ( $= C/T^3 - (\gamma/T^2 + \beta + \sigma T^2)$ ) around 6 K, as shown in ZrTe<sub>3</sub>.

In order to investigate the origin of the  $C_{\text{ex}}$ , we use a modified Debye spectrum for the phase-mode excitations with two cutoff frequencies; the lower one corresponding to the pinning frequency ( $\nu_0 = k_B T_0/h$ ) and the upper one analogous to a Debye temperature for phonons, the "phonon Debye frequency", ( $\nu_\phi = k_B T_\phi/h$ ) [1, 2]. The  $C_{\text{ex}}$  is expressed as

$$C_{\text{ex}} = 3N_0 k_B \left(\frac{T}{T_\phi}\right)^3 \int_{T_0/T}^{T_\phi/T} (x - x_0)^2 \frac{x^2 e^x}{(e^x - 1)^2} dx \quad (1)$$

with  $x = h\nu/k_B T$ ,  $N_\phi$  is the number of phonon mode excitations, and  $k_B$  Boltzmann's constant. The values of the best fit to Eq. (1) are  $N_\phi = 0.32 N_0$  ( $N_0$ , the Avogadro number),  $T_0 = 28$  K (corresponding  $\nu_0 = 580$  GHz), and  $T_\phi = 38$  K for ZrTe<sub>3</sub>, and these values for

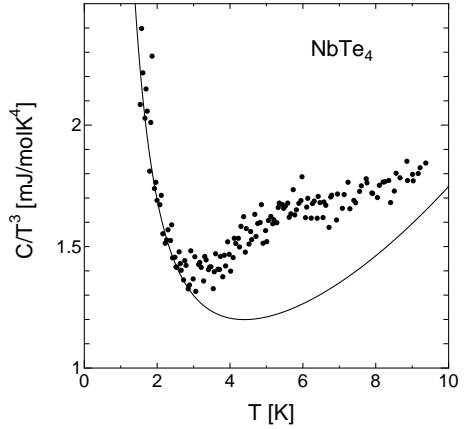


Fig. 2. Temperature dependence of the specific heat of NbTe<sub>4</sub> in a  $C/T^3$  vs.  $T$  plot. Solid line is described by the function of  $C/T^3 = \gamma/T^2 + \beta + \sigma T^2$ .

NbTe<sub>4</sub> are  $N_\phi = 0.18 N_0$ ,  $T_0 = 26$  K (corresponding  $\nu_0 = 540$  GHz), and  $T_\phi = 34$  K. The values of the pinning frequency (540 - 580 GHz) obtained are significantly larger than those of (TaSe<sub>4</sub>)<sub>2</sub>I ( $\nu_0 = 42$  GHz) and KCP ( $\nu_0 = 200$  GHz) where the nonlinear conduction due to the CDW motion is observed [1, 2]. From the relation between the threshold electric field  $E_{\text{th}}$  for the pinned CDW and the pinning frequency,  $E_{\text{th}} = m* \nu_0^2 / 2k_F e$  ( $m*$  the effective mass of electrons,  $k_F$  Fermi wave number and  $e$  electron charge), the value of  $E_{\text{th}}$  for ZrTe<sub>3</sub> and NbTe<sub>4</sub> are estimated as  $10^2$  -  $10^3$  V/cm, which is more than two order of magnitude larger than the values reported in other CDW compounds. This is not inconsistent with the experimental result where the nonlinear conduction is not observed below 10 V/cm in ZrTe<sub>3</sub>. These findings may be related to facts in which the CDW Q-vectors of ZrTe<sub>3</sub> and NbTe<sub>4</sub> are almost commensurate to their lattice [4, 5].

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