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Quasiparticle density of states of $2H$-$NbSe_2$ single crystals revealed by low-temperature specific heat measurements according to a two-component model*

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Low-temperature specific heat in a dichalcogenide superconductor $2H$-$NbSe_2$ is measured in various magnetic fields. It is found that the specific heat can be described very well by a simple model concerning two components corresponding to vortex normal core and ambient superconducting region, separately. For calculating the specific heat outside the vortex core region, we use the Bardeen–Cooper–Schrieffer (BCS) formalism under the assumption of a narrow distribution of the superconducting gaps. The field-dependent vortex core size in the mixed state of $2H$-$NbSe_2$, determined by using this model, can explain the nonlinear field dependence of specific heat coefficient $\gamma(H)$, which is in good agreement with the previous experimental results and more formal calculations. With the high-temperature specific heat data, we can find that, in the multi-band superconductor $2H$-$NbSe_2$, the recovered density of states (or Fermi surface) below T_c under a magnetic field seems not to be gapped again by the charge density wave (CDW) gap, which suggests that the superconducting gap and the CDW gap may open on different Fermi surface sheets.

Keywords: $2H$-$NbSe_2$, charge density wave, superconductivity, specific heat
PACC: 7125, 7145L, 7430E, 7420F

1. Introduction

$2H$-$NbSe_2$, a member of transition metal dichalcogenides, is an interesting superconductor because the electron-phonon coupling leads to the coexistence of the superconductivity (SC) and the charge density wave (CDW). The relationship between SC and CDW is still an issue of long standing. By changing some parameters, for example, by intercalating alkaline elements[1,2] into the parent materials or by changing pressure,[3] it was found that the CDW and the SC could compete each other and a phase diagram looked similar to that of cuprates.[4] Recently experiments by using angle-resolved photo-emission spectroscope (ARPES) showed that the superconducting gap in $2H$-$NbSe_2$ was Fermi surface sheet-dependent.[5,6] Meanwhile, the superconducting state of this system has been studied extensively by specific heat measurement. One of the most interesting issues based on these experiments is the nonlinear field dependence of the electronic specific heat coefficient $\gamma(H)$. It was first suggested that this coefficient was due to the shrinking of the vortex cores.[7] Similarly, Hanaguri *et al* pointed out that the $\gamma(H)$ anomaly was caused by quasiparticle density of states (DOS) outside the vortex cores.[8] The close relationship of the nonlinear $\gamma(H)$ with the delocalized quasiparticles and the vortex core size has been well established from other studies,[9−11] which confirms the microscopic theory for interacting vortices in single-band and two-band systems.[12−14] However, it remains unclear whether the SC and the CDW phases can compete each other, and how the possible competition influences the low energy excitation in this interesting system.

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In this paper, we report the measurements of the specific heat of $2H$-$NbSe_2$ single crystals in various magnetic fields up to H_{c2} and a large temperature region covering the CDW transition. It is found that a two-component model in the framework of Bardeen–Cooper–Schrieffer (BCS) theory can be used to simulate the experimental data very well and vividly present the shrinking of vortex core in $2H$-$NbSe_2$ due to quasiparticle delocalization. By comparing the data of superconducting state and CDW state, we suggest that the superconducting gap and the CDW gap may open on different Fermi surface sheets.

2. Experiment

The samples used in our measurements were single crystals with transition temperatures T_{CDW}=35 K and T_c=7.1 K. They were the selected best samples with dimensions of $3.1 \times 2.8 \times 0.35\,mm^3$ and the steepest superconducting transition to measure the specific heat. The specific heat from 1.8 to 50 K in magnetic fields of zero and 8 T was measured first. And then in a low temperature region from 1.8 to 12 K, the specific heat was measured in magnetic fields varying from 0 to 4 T in steps of 0.5 T. All the measurements were performed with a Quantum Design physical property measurement system (PPMS). The calorimeter puck had been calibrated in magnetic fields to avoid the effect of the thermometer magnetoresistance. The specific heat of the addenda was subtracted automatically by the software through two steps of measurements: first, we measured the specific heat of only the addenda with a little grease on it, and then the specific heat of the addenda together with the sample on it. The magnetic field was applied parallel to the c-axis of the samples in all the measurements. All the data of specific heat were obtained with temperature increasing after the sample had been cooled in a magnetic field from a temperature well above T_c, namely, in a field cooling process.

3. Results and discussion

The specific heat data in a wide temperature range which covers the temperatures of both the CDW and superconducting transitions are shown in Fig.1. The inset gives an enlarged view around the superconducting anomaly. The two transitions take place at T_{CDW} = 35 K and T_c = 7.1 K respectively, both judged by the starting point of the transitions. The anomaly corresponding to the CDW transition is a little wide but clear enough to see. On the contrary, the superconducting transition which occurs against the background of the CDW state is very sharp. At a high field of 8 T, the anomaly corresponding to the CDW transition does not change at all compared with that in a zero field, while the superconducting anomaly is suppressed completely, showing a strong field induced pair breaking effect. A high magnetic field suppresses the superconductivity and recovers a normal state which is exactly the same as the state just above T_c. This would not happen if the field induced extra DOS or the newly emergent Fermi surface segments after using a magnetic field in the superconducting state were gapped again by the CDW phase. In other words, the SC comes from the residual electronic states after the formation of CDW phase, and such residual states will not be further gapped away by the CDW phase with the increase of field below T_c. This suggests that the SC and the CDW gaps may open on different Fermi surface sheets.

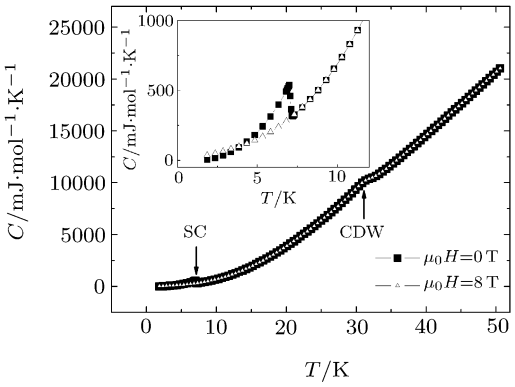


Fig.1. Temperature dependences of specific heat from 1.8 to 50 K at μ_0H=0 and 8 T. The inset is an enlarged view around the superconducting anomaly.

Figure 2 shows the specific heat data as C/T versus T^2 from 1.8 to 10 K with magnetic field increasing from 0 to 4 T in steps of 0.5 T. The critical temperature is suppressed gradually by the magnetic field from 7.1 K to a temperature below 1.8 K. Above 4 T, the sample turns into the normal state entirely. The results are consistent with that given by Sanchez *et al.*[15] From the data at 4 T we find that the normal-state specific heat can be described by $C_n/T = \gamma_n + \beta T^2$. The first term on the right-hand side is the electronic specific heat in the normal state, and the second term is due to the phonon contribution. Fitting to the formula mentioned above,

the coefficients $\gamma_n = 17.67\,mJ{\cdot}mol^{-1}{\cdot}K^{-2}$ and $\beta = 0.51\,mJ{\cdot}mol^{-1}{\cdot}K^{-4}$ are determined. From the relation: $\beta = \frac{12\pi^4}{5}\frac{N_A k_B Z}{\Theta_D^3}$, where $N_A = 6.02 \times 10^{23}$ is the Avogadro constant, and $Z = 3$ the number of atoms in one unit cell, we obtain the Debye temperature $\Theta_D = 225.3\,K$.

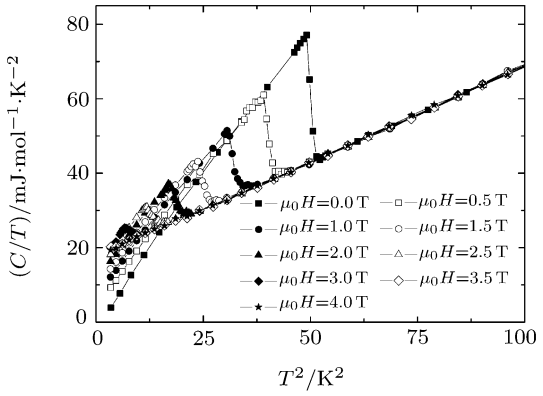


Fig.2. Specific heat in C/T versus T^2 in various fields parallel to the c-axis of the sample.

By subtracting the phonon contribution the electronic specific heat C_e is obtained. In the superconducting state, $C_e = \gamma T$, where γ is the electronic specific heat coefficient that is dependent on temperature and field. In Fig.3 the value of γ in a zero field, subtracted by the value o γ_n, is plotted, and the solid line denotes a theoretical fit that will be described in detail in the following section. From this figure we find $\Delta C/\gamma_n T\,|_{T_c} = 1.955$. This value is larger than the weak-coupling limit BCS value 1.43, which may indicate that this system belongs to the moderate coupling case.

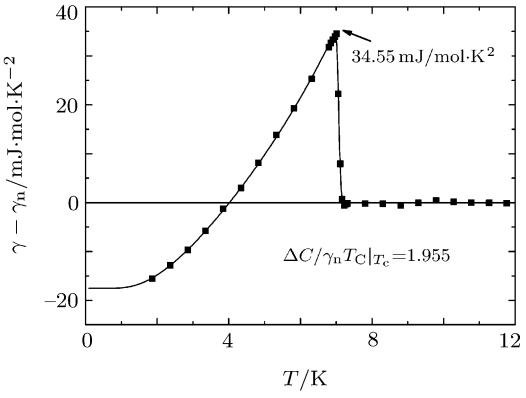


Fig.3. Temperature dependence of the electronic specific heat coefficient in a superconducting state under a zero field. The solid line represents a theoretical fit to expression (2).

The upper critical field determined by the midpoints of superconducting anomaly of the specific heat is shown in Fig.4. The solid line denotes a fit by using the slightly modified formula based on the Ginzburg–Landau theory

$$H_{c2} = H_{c2}(0)(1 - t^2)/(1 + at^2), \qquad (1)$$

where $t = T/T_c$, and $a = 0.4$. It fits the experimental data fairly well, leading to the determination of the zero-temperature upper critical field $\mu_0 H_{c2}(0) = 3.68 \pm 0.1\,T$.

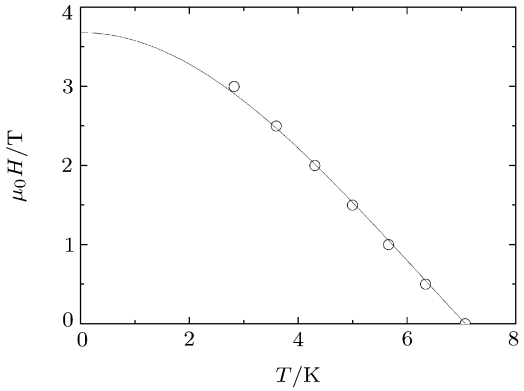


Fig.4. The upper critical fields determined by the midpoints of the jump of the specific heat, with the solid line denoting a fit by using expression (1).

Next we show the quantitative analysis on the specific heat data both in a zero field and in a mixed state. We use the formula which is derived from thermodynamic relations based on the BCS theory[16] for calculating the specific heat. The electronic specific heat in the zero field can be written as

$$C_{es} = \frac{4N(0)}{k_B T^2}\int_0^\infty \frac{1}{2\pi}\int_0^{2\pi} \frac{e^{\zeta/k_B T}}{(1+e^{\zeta/k_B T})^2} \times \left(\varepsilon^2 + \Delta^2 - \frac{T}{2}\frac{d\Delta^2}{dT}\right) d\varepsilon\, d\theta, \qquad (2)$$

where $\zeta = \sqrt{\varepsilon^2 + \Delta^2}$ and $\Delta = \Delta(T)f(\theta)$, with $\Delta(T)$ being the temperature dependent BCS gap function. Neither the isotropic gap ($f(\theta)$=1) nor the gap with nodes (e.g. $f(\theta) = |\cos 3\theta|$) could fit the experimental data. On the contrary, an anisotropic gap or two isotropic gaps can fit the data very well, which is shown in Fig.5 as solid lines for $H = 0$ according to an anisotropic gap (a) and two isotropic gaps (b). For the anisotropic gap, $f(\theta) = 0.4 + 0.6|\cos 3\theta|$, and the gap value at a zero temperature is $\Delta_0 = 1.62\,meV$ [i.e. $\Delta(T = 0)$]. Here we have chosen $|\cos 3\theta|$ because $2H$-$NbSe_2$ has a triangular lattice structure, which may lead to 3- or 6-fold symmetry as suggested in Ref.[4]. We have also tried to use other form of gap with a similar level of anisotropy mentioned in other more formal calculations,[13] to do the same job. It is found

that the fitting curves are sensitive only to the level of the anisotropy but not to the selected form of gap function. For the two-gap model, the gap values at a zero temperature are $\Delta_1 = 1.50\,\mathrm{meV}$, $\Delta_2 = 0.85\,\mathrm{meV}$ and the two parts with different gap values separately occupy 64% and 36% in the whole superconducting electronic specific heat. Both the anisotropic and the two-gap model can fit the data very well.

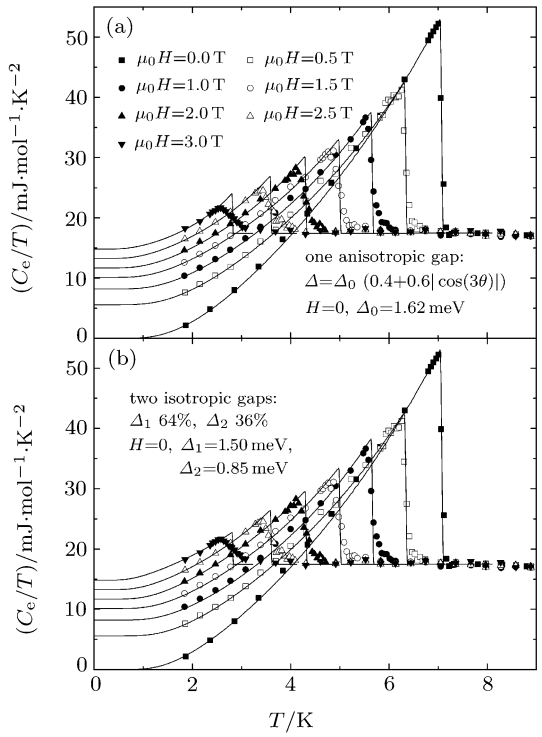


Fig.5. The calculation result by using the two-component model according to an anisotropic gap (a) and two isotropic gaps (b).

In the mixed state, there are two different regions, namely the core region and the outside core region. Therefore we adopt an improved two-component model[17,18] which separates the electronic specific heat into two components. The electronic specific heat is thus written as

$$C_{\mathrm{e}} = \alpha\frac{H}{H_{\mathrm{c2}}(0)}\gamma_{\mathrm{n}}T + \left(1-\alpha\frac{H}{H_{\mathrm{c2}}(0)}\right)C_{\mathrm{es}}. \quad (3)$$

The first term on the right-hand side is the density of states coming from the normal vortex core regions, and the second part originates from the superconducting regions outside the cores. Recently, Shan *et al* [18] used this two-channel model to explain the point-contact Andreev reflection spectra measured in various magnetic fields successfully. In this model, the parameter α was introduced to take into account the vortex shrinking caused by any mechanism. Moreover, $\alpha H/H_{\mathrm{c2}}(0)\gamma_{\mathrm{n}}$ is obviously equal to the zero-temperature electronic specific heat coefficient $\gamma(H)$ according to expressions (2) and (3).

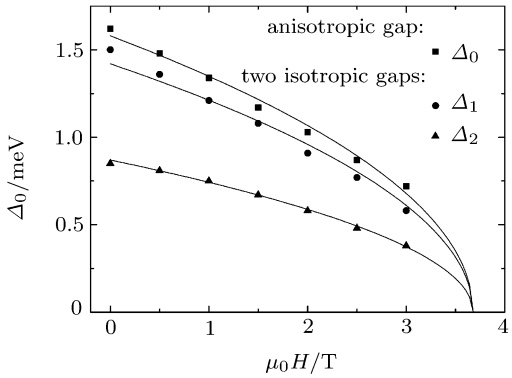


Fig.6. Field dependences of the gap value. The solid squares are denote the calculations obtained by using the anisotropic gap. The dots and triangles refer to the results obtained from the two-gap model. The lines represent the relation $\Delta \propto \sqrt{1-H/H_{\mathrm{c2}}}$[25–27] based on the figure of magnetic field induced pair breaking varying with H/H_{c2} as the pair breaking factor.

The fitting results according to the two-component model for the mixed state are shown in Fig.5. The anisotropic gap and the two-gap model almost bring the same results both in a zero field and the mixed state, and the determined values of $\gamma(H)$ are also identical. We could not distinguish the gap forms by using the fitting to specific data both in a zero field and the mixed state. According to these results, we demonstrate the strong k-dependence of the superconducting gap. The band structure of $2H$-$NbSe_2$ consists of quasi-2D sheets and a 3D 'pancake-like' pocket.[19] Several measurements have shown the evidence for gap anisotropy in this system.[9,20–22] Yokaya *et al* [5] measured the SC gap at different points in k-space. Their results suggested no gap or a very small one in the small 3D pocket if it even exists, and gaps with different values on the two quasi-2D sheets. Since the density of states on the 3D sheet is too small, we think that the anisotropy of the gaps mostly arises from the quasi-2D sheets. However, we cannot distinguish the two possible cases from each other: multi-gap model and a single anisotropic gap. This is in good agreement with the recent measurements of the penetration depth and specific heat.[21,23] A recent work on the Al-doped MgB_2[24] has pointed out that the evolution of the gaps suggests that the interband scattering increases with Al content increasing. Therefore, for $NbSe_2$, if the gaps on the two 2D fermi surfaces are similar to each other, the interband

scattering may smear up the two gap effect and become similar to the case of single anisotropic gap. The field dependence of the average zero-temperature gap values is plotted in Fig.6. The solid lines show the relation $\Delta(T=0) \propto \sqrt{1-H/H_{c2}}$[25−27] which is derived for the magnetic field induced pair breaking varying with H/H_{c2} as the average pair breaking factor.

In what follows, we will testify the validity of the above analysis by comparing the physical quantities derived here with those determined by other groups or by using other experimental methods. Figure 7 shows the field dependence of the electronic specific heat coefficient $\gamma(H)$ at a zero temperature, determined from our general fit. As one can see, the determined $\gamma(H)$ in our work is in good agreement with those obtained by other groups,[7,15,28] and a low field nonlinearity is very clear. There are two possible reasons for interpreting this nonlinearity: multigap effect and delocalization of quasiparticles.[7,29] In MgB_2 a strong nonlinear effect of $\gamma(H)$ has been observed due to the easy pair breaking by magnetic field from the π-band with a much small gap compared with the σ-band gap. As we stated already, there are indeed multigaps in $2H$-$NbSe_2$, but the total density of states arising from the small pancake-like Fermi surface around the Γ point is very small, this little contribution is certainly not enough to give rise to a nonlinear effect on the $\gamma(H)$ relation. Furthermore, based on the fitting results, we have shown that, even though there are two sets of cylinder-like Fermi surfaces, the gaps on them are quite close to each other, which would be also difficult to bring about the nonlinear effect on $\gamma(H)$. In this

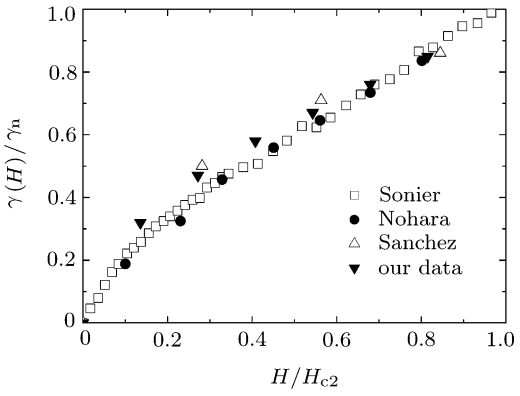


Fig.7. The electronic specific heat coefficient $\gamma(H)$ normalized to the normal state value vs H/H_{c2}. The solid triangle denotes our data determined from the theoretical calculations based on the two-component model. The others are for the data given by Sonier *et al*[7] at $T = 2.3$ K by changing the field, Nohara *et al*[28] through extrapolating to $T \to 0$, and Sanchez *et al*[15] through using the two-component model

case, the nonlinear field dependence of $\gamma(H)$ may be naturally attributed to the delocalization of the quasiparticle states. It is noted that $C(T)$ may have a T^2-term in the vortex state arising from the V-shape DOS at the Fermi surface as pointed out by Nakai *et al.*[30] Our simple model cannot reflect such a T^2-term. However, if it indeed exists, the obtained $\gamma(H)$ will be modified slightly due to the small slope of T^2 term on the C/T versus T plot and the constrain of entropy conservation.

In addition, we find that the two-component model can fit the experimental data very well, with only a little modification required. From the data shown in Fig.5, it is seen that the jump of the specific anomaly becomes wider and wider with magnetic field increasing, but the calculations cannot fit that in tail. It is thus assumed that the gap has a slight distribution with $G(\Delta)$ as a distribution function. We further assume that $G(\Delta)$ has a form of a Gaussian distribution

$$G(\Delta) = \frac{1}{w\sqrt{\pi/2}} \mathrm{e}^{-2(\frac{\Delta-\Delta_0}{w})^2}, \qquad (4)$$

where Δ_0 means $\Delta_{BCS}(T)$, and w is the full width at half-maximum (FWHM) of the gap distribution function.

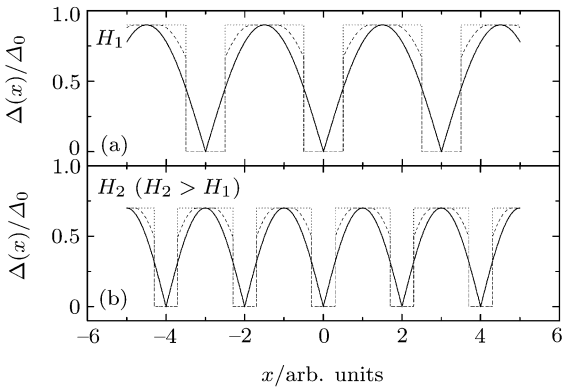


Fig.8. Spatial variations of the normalized order parameter, separately, in the presence of Abrikosov vortices, shown by solid lines, and in two-component model, shown by dotted lines for low field (a) and high field (b). Where the dashed lines are for the two-component model with a gap distribution.

The reason why the gap distribution is considered here is that the two-component model is a rigid one while in the real case the normal core and outside region should not have a clear-cut boundary. A schematic plot for the spatial distribution as a function of gap value in the mixed state is shown in Fig.8 as the solid lines. The dotted lines show the situation for the rigid two-component model, and dashed lines

are for the case in which the gap distribution in taken into account. For the low field case [Fig.8(a)], the vortex cores are located far from each other, and Fig.8(b) shows the case for a higher field. The distance between the cores decreases with the increase of magnetic field. Since the spatial distribution of the order parameter or the gap values is closely related to magnetic field, we naturally modify the rigid model by treating this effect as a gap with the Gaussian distribution. An integral of gap is carried out and expression (2) becomes

$$C_{\rm es} = \frac{4N(0)}{k_{\rm B}T^2}\int_{-\infty}^{\infty}\frac{1}{2\pi}\int_0^{2\pi}\int_0^{\infty}\frac{{\rm e}^{\zeta/k_{\rm B}T}}{(1+{\rm e}^{\zeta/k_{\rm B}T})^2} \times\left(\varepsilon^2+\Delta^2-\frac{T}{2}\frac{{\rm d}\Delta^2}{{\rm d}T}\right)G(\Delta)\,{\rm d}\Delta\,{\rm d}\theta\,{\rm d}\varepsilon. \quad (5)$$

The modified two-component model with the Gaussian gap distribution can fit the experimental data very well, which is shown in Fig.9. Only a very small FWHM of the distribution is needed (from 0.015 for $\mu_0 H = 0\,{\rm T}$ to 0.075 for $\mu_0 H = 3\,{\rm T}$), indicating that the original rigid model may have already provided a good basis. In addition, at a zero field there occurs also a little broadening of the transition. This may be attributed to the slight contribution of the defects in the sample, which can also be treated by a Gaussian gap distribution with a small FWHM. This two-component model can reproduce the results from other groups very well.[15,29] For the data of Nb, reported by Sonier *et al*,[29] the two-component model with an isotropic gap function works very well, indicating a similar low energy excitation in these two different materials. And the data of $NbSe_2$, reported by Sanchez, encounter the same instance as ours.

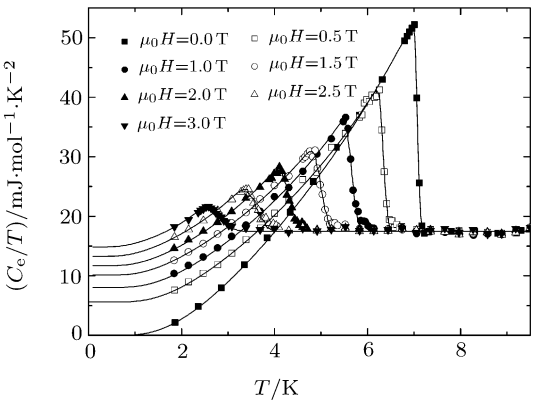


Fig.9. $C_{\rm e}/T$ versus T, where the lines represent theoretical calculations obtained from the revised two-component model.

4. Conclusions

Through the measurements of low temperature specific heat of $2H$-$NbSe_2$, we find that the recovered density of states (or Fermi surface) below $T_{\rm c}$ under a magnetic field seems not to be gapped again by the charge density wave (CDW) gap, which suggests that the superconducting gap and the CDW gap may open on different Fermi surface sheets. For the superconducting state, we use a two-component model based on the BCS theory to fit the experimental data. The form of the superconducting gap cannot be determined through the fitting, for both the anisotropic gap model and two-gap model fit the data well. The determined $\gamma(H)$ at a zero temperature shows its nonlinearity with H, which is consistent with the previous work. In addition, the data can be fitted very well by the modified two-component model with a narrow distribution of the superconducting gaps outside the vortex cores.

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