Temperature dependence of the critical field of the organic superconductor κ -(BEDT-TTF)₂Cu[N(CN)₂]Br

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We present upper critical magnetic fields data for κ -(BEDTTTF)₂Cu[N(CN)₂]Br (hereafter k-Br), single crystals with magnetic field applied either perpendicular or parallel to the conducting layers. H-T phase diagram in this compound has been deduced from tunnel diode oscillator-based contactless measurements in pulsed magnetic fields up to 56 T for the inter-plane (H||b) and in-plane (H||ac) field directions. Temperature dependence of the magnetic penetration depth in DC fields is also reported. The temperature dependence of the upper critical magnetic field H_{c2} is not accounted for the Werthamer–Helfand–Hohenberg (WHH) model for in-plane configuration. For the inter-plane orientation, a significant upward curvature is observed as temperature decrease, in the range close to T_c , followed by saturation at lower temperatures. Possibly, this upturn is an indication of flux-line lattice melting.

The temperature-dependence of the upper critical magnetic field $H_{c2}(T)$ in organic superconductors is most likely due to two independent pair-breaking mechanism [1,2]: (i) close to T_c . Cooper pairing is suppressed by orbital currents that screen the external field, according to the well-known WHH model; (ii) towards lower temperatures, the limiting effect is caused by the Zeeman splitting, i.e., when the Zeeman energy becomes larger than the condensation energy, the Pauli limit, H_p , is reached. This paramagnetic limit, $H_{c2}(T)$, is lower than the orbital one which is related to the slope $dH_{c2}(T)/dT$ close to T_c . Namely, when including Pauli paramagnetism, the upper critical field is reduced relatively to $H_{c2}(T)$:

$$H_{c2}(T) = H_{c2}^{*}(T) / \sqrt{1 + \alpha^{2}(T)}, \qquad (1)$$



Fig. 1. (Color online) Phase diagram for the k-Br compound with conducting plane oriented parallel (H||ac) and perpendicular (H||b) to the pulsed magnetic fields. Black and blue squares indicate the H_{c2} values inferred from experiments with H||ac and H||b, respectively. Green squares correspond to the melting transition fields $H_m(T)$ data. The red dotted line is the best fit of the WHH model to the data for H||ac. Green dotted line is power low fit close to T_c . [1, 5, 6]. The red dotted line indicate the temperature dependence according to the WHH model neglecting Pauli-limiting

where $\alpha(T) = \sqrt{2}H_{c2}/H_p(0)$ is the Maki parameter [3]. Whether a superconductor is orbitally limited or Pauli limited can be inferred from the α parameter value. Above H_p the Cooper pairs are broken and superconductivity is destroyed. The superconductor made up of weakly-coupled superconducting planes [4] may transform from a three-dimensional system to two-dimensional one [5]. Recently, the change in gra-

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dient of H_{c2} vs T in [5] was attributed to dimensional crossover from quasi-two-dimensional (low temperatures) to three dimensional (high temperatures). In this short communication, we report (i) results of the magnetic penetration depth measurements in zero-field for κ -(BEDTTTF)₂Cu[N(CN)₂]Br single crystals in the temperature range between 1.2 and 15 K and (ii) critical magnetic fields measurements in pulsed fields of up to 56 T in the range 1.2 to 12 K. In both cases, field directions parallel and perpendicular to the conducting ac plane were explored.

Here, we found that below 9 K a definite change in the slope of H_{c2} vs T occurs and H_{c2} begins to follow the power law, $H_{c2}(T) \propto (9.8 - T^*)^4$, with fitted $T^* = 9.8$ K. Notice that this upturn temperature position is almost the same as for $H_m(T)$. Similar upturn was observed for λ -(BETS)₂GaCl₄ organic superconductor, with T^* as a free parameter. Following [5], we attribute the sharp upturn of Hc2(T) to flux-line lattice melting, and thus characteristic of a two-dimensional superconductor with weakly-coupled layers, because H^{*} follows the power like dependence $H_{c2} \propto (9.8 - T)^4$ dependence, indeed expected from the Gorter–Casimir two-fluid model [6]. Measurements of the temperature dependence of both upper critical magnetic field and melting transition reveal unusual phase diagram for κ -(BEDT-TTF)₂Cu[N(CN)₂]Br organic superconductor. The temperature dependence of the upper critical magnetic field $H_{c2}(T)$ shows an upward divergence as the temperature decreases with a sharp upturn at 9 K, and similar upturn for the temperature dependence of melting field $H_m(T)$ (see Fig. 1). We tentatively attribute these two upturns to flux-line lattice melting.

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- J. Singleton and Ch. Mielke, Contemp. Phys. 43, 63 (2010).
- G. Zwicknagl and J. Wosnitza, Int. J. Mod. Phys. B 24, 3915 (2010).
- 3. K. Maki, Phys. Rev. **148**, 362 (1966).
- 4. L. Bulaevskii, Adv. Phys. 37, 443 (1988).
- Ch. Mielke, J. Singleton, M.-S. Nam, N. Harrison, C. C. Agosta, B. Fravel, and L. K. Montgomery, J. Phys.: Cond. Matt. 13(36), L203 (2001).
- C. J. Gorter and H. B. G. Casimir, Phys. Z. 35, 963 (1934).