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# Vortex state of topological superconductor $\text{Cu}_x\text{Bi}_2\text{Se}_3$

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The electron pairing in the topological superconductor has been predicted to be spin-triplet pairing with odd parity. If the superconducting carries have a magnetic moment, the magnetization arising from it reduces the repulsive interaction between the vortices, thus the vortex density drastically increases in the region above the lower critical magnetic field  $H_{c1}$ . The comparison of the theoretical calculation with the experimental result by Das *et al.* [*Phys. Rev. B* **83**, 220513(R) (2011)] indicates that the magnetization is small, suggesting that it is not a spin-triplet superconductor. The theory suggests that if the nonzero magnetization arises from the supercurrent carriers a hysteresis in the magnetization curve around  $H_{c1}$  may be observed. The observation (or nonobservation) of it will narrow down the mechanism of the superconductivity in  $\text{Cu}_x\text{Bi}_2\text{Se}_3$ .

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## I. INTRODUCTION

The crystal of  $\text{Bi}_2\text{Se}_3$  is a topological insulator [1]. Upon Cu doping, it becomes a superconductor [2]. The Cu ions are intercalated between the van der Waals layers of Se ions in the crystal, and the domains intercalated by the Cu ions become superconducting [2]. Fu and Berg have theoretically predicted that the superconductivity of  $\text{Cu}_x\text{Bi}_2\text{Se}_3$  is a spin-triplet pairing superconductor with odd parity due to the strong spin-orbit interaction [3]. On the other hand, the specific heat measurement [4] and the STM measurement [5] indicate that the energy gap is in accordance with the *s*-wave pairing, suggesting it is not the triplet pairing.

Das *et al.* made the cylindrical-shaped single crystal (diameter 6 mm and height 1 mm) of  $\text{Cu}_{0.15}\text{Bi}_2\text{Se}_3$  [6]. The superconducting transitions of the sample occurs at the temperature of 3.6 K [6]. They measured the DC magnetization of the crystal with the use of a SQUID magnetometer (Quantum Design Inc., USA). The superconducting state of the crystal shows an intriguing behavior in the vortex state just above the lower critical magnetic field  $H_{c1}$ . When an external magnetic field above  $H_{c1}$  is applied, the vortices enter into the superconducting domains and the magnetization sharply increases just above  $H_{c1}$ . When the magnetic field further increases, the magnetization gradually increases and the magnetization curve finally merges with the normal state diamagnetic curve at  $H_{c2}$ . The measured  $H_{c1}$  for  $\text{Cu}_{0.15}\text{Bi}_2\text{Se}_3$  is about 5 Oe, and magnetization  $4\pi M$  at  $H_{c1}$  is about  $-0.025$  gauss, indicating the volume fraction of the superconducting domains is  $5 \times 10^{-3}$ , which is unusually small.

If the magnetization of the triplet pairs exists, the sharp increase of the magnetization curve just above  $H_{c1}$  is expected, giving very different behavior from the magnetization curves of usual type II superconductors. To clarify whether such a magnetization exists or not, we calculate the magnetization curve above the critical magnetic field  $H_{c1}$  in the superconducting state of  $\text{Cu}_x\text{Bi}_2\text{Se}_3$ . We show in the present paper

that the magnetization is actually small, suggesting that it is not a spin-triplet superconductor. It is also indicated that if the magnetization exists, the magnetization curve may show a hysteresis around  $H_{c1}$  due to the double valuedness of  $M$  as a function of  $H$ .

Considering the similarity between  $\text{Cu}_x\text{Bi}_2\text{Se}_3$  and the cuprate superconductor, the recently proposed superconductivity mechanism based on the spin-vortex formation may be relevant [7–11]. This mechanism will be realized if spin vortices are formed in the real space with the doped coppers as their cores. A slight magnetization will arise from the spin vortices; then, the above mentioned hysteresis may be observed.

## II. FORMALISM

Let us discuss the electromagnetism in the superconducting domains created by the Cu ions intercalated between the Se atomic layers. The magnetic induction  $\mathbf{b}$  in the vortex is given by a sum of the magnetic field  $\mathbf{h}$  induced by the vortex current and the spin magnetization  $\mathbf{m}$  induced by the vortex magnetic field as shown,

$$\mathbf{b} = \mathbf{h} + 4\pi\mathbf{m}. \quad (1)$$

The Maxwell equation is given by

$$\nabla \times \mathbf{h} = \frac{4\pi}{c} \mathbf{j} + \frac{1}{c} \frac{\partial}{\partial t} \mathbf{e}, \quad (2)$$

where  $\mathbf{j}$  is the electric current,  $\mathbf{e}$  is the electric field, and  $c$  is the light velocity. Introducing the vector potential  $\mathbf{a}$ , we express  $\mathbf{e}$  and  $\mathbf{b}$  as

$$\mathbf{e} = -\frac{1}{c} \frac{\partial}{\partial t} \mathbf{a}, \quad (3)$$

$$\mathbf{b} = \nabla \times \mathbf{a}.$$

The current  $\mathbf{j}$  in Eq. (2) is given by Eq. (2.5) in Ref. [12] as

$$\mathbf{j} = \frac{c^2 \hbar}{4\pi \lambda^2 e} \int d^3 r' c^{(3)}(\mathbf{r} - \mathbf{r}') \left[ \nabla_{\mathbf{r}'} f(\mathbf{r}') - \frac{e}{c \hbar} \mathbf{a}(\mathbf{r}') \right], \quad (4)$$

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were  $\lambda$  is the London penetration depth,  $e$  is the absolute value of the electron charge, and  $\hbar$  is the Planck constant divided by  $2\pi$ . The function  $f(\mathbf{r}')$  in Eq. (4) is equal to half the phase of the superconducting order parameter, and  $c^{(3)}(\mathbf{r}-\mathbf{r}')$  is the boson characteristic function [12]. If we take  $c^{(3)}(\mathbf{r}-\mathbf{r}')$  to be the three dimensional delta function  $\delta^{(3)}(\mathbf{r}-\mathbf{r}')$ , Eq. (4) becomes the London equation. The magnetization  $\mathbf{m}$  in Eq. (1) is given by

$$\mathbf{m}(\mathbf{r}) = \chi \mathbf{h}(\mathbf{r}), \quad (5)$$

$\chi$  being the spin magnetic susceptibility of the spin triplet superconductor. For a single vortex,  $f(\mathbf{r}')$  in Eq. (4) fulfills the equation

$$\nabla_{\mathbf{r}'} \times \nabla_{\mathbf{r}'} f(\mathbf{r}') = \pi \mathbf{e}_3 \delta^{(2)}(\mathbf{r}'), \quad (6)$$

$\mathbf{e}_3$  being the unit vector in the third direction. Applying the curl operator  $\nabla_{\mathbf{r}} \times$  on both sides of Eq. (1) and using Eqs. (2)–(5), we obtain the following equation:

$$\begin{aligned} \nabla_{\mathbf{r}} \times \mathbf{b}(\mathbf{r}) &= \frac{c\hbar}{\lambda^2 e} \int d^3 r' c^{(3)}(\mathbf{r}-\mathbf{r}') \left[ \nabla_{\mathbf{r}'} f(\mathbf{r}') - \frac{e}{c\hbar} \mathbf{a}(\mathbf{r}') \right] \\ &\quad - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{a}(\mathbf{r}) + 4\pi \chi \nabla_{\mathbf{r}} \times \mathbf{h}(\mathbf{r}). \end{aligned} \quad (7)$$

Applying the curl operator on both sides of Eq. (7), we have

$$\begin{aligned} \nabla_{\mathbf{r}} \times [\nabla_{\mathbf{r}} \times \mathbf{b}(\mathbf{r})] &= \frac{c\hbar}{\lambda^2 e} \int d^3 r' c^{(3)}(\mathbf{r}-\mathbf{r}') \left[ \nabla_{\mathbf{r}'} \times \nabla_{\mathbf{r}'} f(\mathbf{r}') \right. \\ &\quad \left. - \frac{e}{c\hbar} \mathbf{b}(\mathbf{r}') \right] + 4\pi \chi \nabla_{\mathbf{r}} \times [\nabla_{\mathbf{r}} \times \mathbf{h}(\mathbf{r})]. \end{aligned} \quad (8)$$

Here we have used the following equality:

$$\begin{aligned} \nabla_{\mathbf{r}} \times \int d^3 r' c^{(3)}(\mathbf{r}-\mathbf{r}') \mathbf{C}(\mathbf{r}') &= - \int d^3 r' \nabla_{\mathbf{r}'} \times [c^{(3)}(\mathbf{r}-\mathbf{r}') \mathbf{C}(\mathbf{r}')] \\ &\quad + \int d^3 r' c^{(3)}(\mathbf{r}-\mathbf{r}') \nabla_{\mathbf{r}'} \times \mathbf{C}(\mathbf{r}') \\ &= \int d^3 r' c^{(3)}(\mathbf{r}-\mathbf{r}') \nabla_{\mathbf{r}'} \times \mathbf{C}(\mathbf{r}'), \end{aligned} \quad (9)$$

where  $c^{(3)}(\mathbf{r}-\mathbf{r}') \mathbf{C}(\mathbf{r}')$  is assumed to vanish on the surface of the integration volume.

The quantities  $\mathbf{h}$  and  $\mathbf{b}$  are parallel to the third axis, thus, we may write  $\mathbf{h} = h\mathbf{e}_3$  and  $\mathbf{b} = b\mathbf{e}_3$ . By assuming the uniformity in the third direction, Eq. (8) is calculated as

$$\begin{aligned} \nabla_{\mathbf{r}}^2 b(\mathbf{r}) &= -\frac{\phi}{\lambda^2} c(\mathbf{r}) + \frac{1}{\lambda^2} \int d^2 r' c(\mathbf{r}-\mathbf{r}') b(\mathbf{r}') \\ &\quad + 4\pi \chi \nabla_{\mathbf{r}}^2 h(\mathbf{r}), \end{aligned} \quad (10)$$

where  $\phi$  is the magnetic flux quantum given by  $hc/2e$ ,  $h$  being the Planck constant, and  $c(\mathbf{r})$  is the two-dimensional version of  $c^{(3)}(\mathbf{r})$  which becomes the two-dimensional delta function  $\delta^{(2)}(\mathbf{r})$  in the London limit. The Fourier transform of

a function  $g(\mathbf{r})$  is expressed as

$$g(\mathbf{r}) = \frac{1}{(2\pi)^2} \int d^2 k g_{\mathbf{k}} e^{i\mathbf{k}\cdot\mathbf{r}}, \quad (11)$$

where the uniformity of the system along the third direction is assumed, i.e.,  $\mathbf{k}$  and  $\mathbf{r}$  are taken as vectors in the plane perpendicular to the third direction.

According to Eq. (11), the Fourier transform of Eq. (10) is obtained as

$$\mathbf{k}^2 b_{\mathbf{k}} = \frac{\phi}{\lambda^2} c_{\mathbf{k}} - \frac{1}{\lambda^2} c_{\mathbf{k}} b_{\mathbf{k}} + 4\pi \chi \mathbf{k}^2 h_{\mathbf{k}}. \quad (12)$$

From Eqs. (1) and (5), we have

$$b_{\mathbf{k}} = (1 + 4\pi \chi) h_{\mathbf{k}}. \quad (13)$$

Then, using Eqs. (12) and (13), we obtain

$$\mathbf{k}^2 (1 + 4\pi \chi) h_{\mathbf{k}} = \frac{\phi}{\lambda^2} c_{\mathbf{k}} - \frac{1}{\lambda^2} c_{\mathbf{k}} (1 + 4\pi \chi) h_{\mathbf{k}} + 4\pi \chi \mathbf{k}^2 h_{\mathbf{k}}. \quad (14)$$

Thus, the Fourier component  $h_{\mathbf{k}}$  of  $h(\mathbf{r})$  is obtained as

$$h_{\mathbf{k}} = \frac{\phi c_{\mathbf{k}}}{(\lambda \mathbf{k})^2 + c_{\mathbf{k}} (1 + 4\pi \chi)}. \quad (15)$$

Finally,  $h(\mathbf{r})$  is obtained, using Eqs. (11) and (15), as

$$\begin{aligned} h(\mathbf{r}) &= \frac{\phi}{(2\pi)^2} \int d^2 k e^{i\mathbf{k}\cdot\mathbf{r}} \frac{c(\lambda k)}{(\lambda k)^2 + c(\lambda k)(1 + 4\pi \chi)} \\ &= \frac{\phi}{2\pi} \int k dk J_0(kr) \frac{c(\lambda k)}{(\lambda k)^2 + c(\lambda k)(1 + 4\pi \chi)}, \end{aligned} \quad (16)$$

where  $J_0(kr)$  is the 0th order Bessel function;  $c_{\mathbf{k}}$  is expressed as  $c(\lambda k)$  assuming that its dependence on  $\mathbf{k}$  is through the product  $\lambda k$ . Actually, we adopt the following form for  $c(\lambda k)$ ,

$$c(\lambda k) = \exp(-\alpha \lambda^2 k^2) \quad (17)$$

This function describes the extent of the current flow around the core of the vortex, assuming that the penetration depth  $\lambda$  is much larger than the size of the vortex core.

The function  $h(\mathbf{r})$  in Eq. (16) depends  $\mathbf{r}$  through  $r$ , thus, we express it as  $h(r)$ . The interaction energy between the vortices whose centers are respectively at the origin  $r = 0$  and the positive  $r$  is given by

$$\frac{\phi}{8\pi} h(r). \quad (18)$$

We calculate the  $r$  dependence of  $h(r)$  for the several values of the spin magnetic susceptibility  $\chi$ . The result is shown in Fig. 1. It shows that when the magnetization arises ( $\chi \neq 0$ ) the repulsive interaction between the vortices is reduced. Actually, even an attractive interaction region with  $h(r) < 0$  exists. This  $\chi$  dependence of  $h(r)$  influences the magnetization curve in the vortex state as will be shown below. By using this  $\chi$  dependence of the magnetization curve, we can estimate the value of  $\chi$ . Since the triplet pairing yields  $\chi \neq 0$ , the occurrence of the spin-triplet pairing will be deduced from the experimentally observed magnetization curve.

When the magnetic field  $H$  larger than  $H_{c1}$  is applied along the third axis of the crystal, the cylindrical vortices parallel to

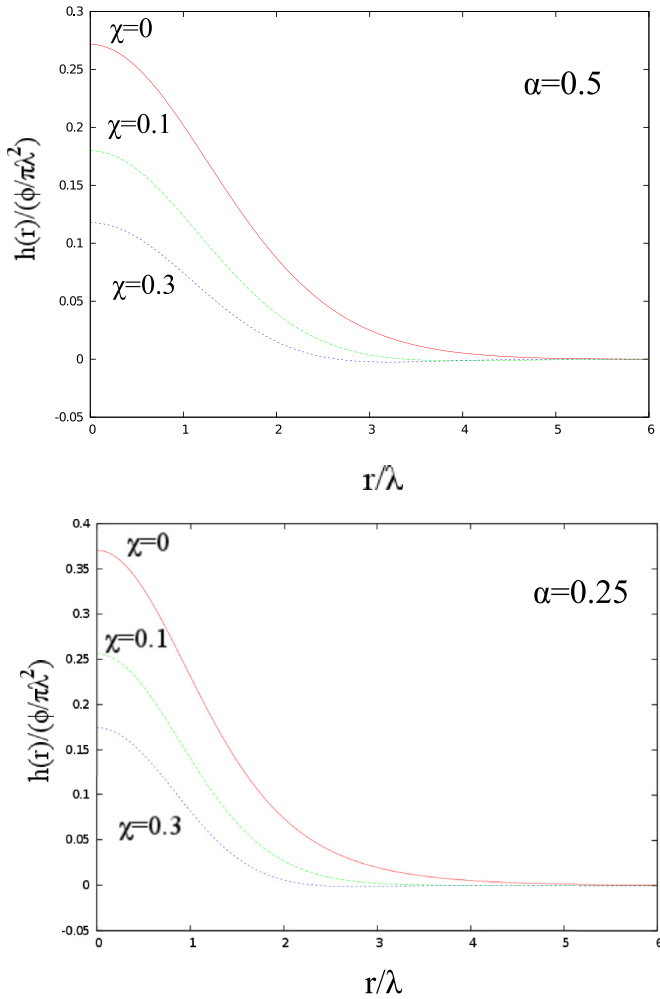


FIG. 1. (Color online) Plots of  $h(r)$  with two different values of  $\alpha$  (0.5 and 0.25). Values of  $\chi$  are 0, 0.1, 0.3 from the top to the bottom.

the magnetic field are generated in the crystal. The Gibbs free energy for the system in this case is given by

$$G = W_{\text{vortex}} - \frac{1}{4\pi} HB. \quad (19)$$

The magnetic induction  $B$  is equal to  $n\phi$  with  $n$  being the vortex number in the unit area. The vortex energy density  $W_{\text{vortex}}$  in Eq. (19) is given by

$$W_{\text{vortex}} = W_{\text{self}} + W_{\text{interaction}}. \quad (20)$$

The self-energy density  $W_{\text{self}}$  is given by the sum of the vortex magnetic self-energy density  $W_{\text{magnetic-self}}$  and the vortex core energy density  $E_{\text{core}}$ , and thus, the self-energy density is expressed as

$$W_{\text{self}} = W_{\text{magnetic-self}} + E_{\text{core}} = n \frac{\phi}{4\pi} H_{c1}^0, \quad (21)$$

where  $\frac{\phi}{4\pi} H_{c1}^0$  is the cost of energy for the creation of a single vortex. The vortex interaction energy is expressed as

$$W_{\text{interaction}} = \frac{\phi}{4\pi} \sum_{i \neq j} h(|\mathbf{r}_i - \mathbf{r}_j|), \quad (22)$$

where  $h(|\mathbf{r}_i - \mathbf{r}_j|)$  is the magnetic field given by Eq. (16), and  $\mathbf{r}_i$  and  $\mathbf{r}_j$  indicate the vortex center positions of the  $i$ th and  $j$ th vortices, respectively. In the present theory, the lower critical field  $H_{c1}$  may be different from  $H_{c1}^0$  due to the existence of the attractive interaction distance between vortices (i.e.,  $h(r) < 0$ ) as seen in Fig. 1 for the  $\chi \neq 0$  cases.

The vortices construct their triangular lattice in the plane perpendicular to the applied magnetic field. When the vortex density is not so large, the interaction strength between the two vortices in the nearest neighbor vortices pair is much stronger than the interactions of the other pairs. Therefore, in this case, we take account of only the interaction energy between the nearest neighbor vortices pair. The length of the nearest neighbor vortex pair is given by  $\sqrt{2}/(\sqrt{3}n)$  and the number of the nearest neighbor vortex pair in the unit area is given by  $3n$  with  $n$  being the vortex density. In this case, the vortex interaction energy density is expressed as

$$W_{\text{interaction}} = \frac{\phi}{4\pi} 3nh(\sqrt{2}/(\sqrt{3}n)). \quad (23)$$

Taking account of Eqs. (20), (21), and (23), the Gibbs free energy Eq. (19) is expressed as

$$G = -n \frac{\phi}{4\pi} (H - H_{c1}^0) + \frac{\phi}{4\pi} 3nh(\sqrt{2}/(\sqrt{3}n)). \quad (24)$$

We obtain the relation between the magnetic field  $H - H_{c1}^0$  and the vortex density  $n$  by the condition  $\partial G/\partial n = 0$ , which yields

$$H - H_{c1}^0 = 3h(\sqrt{2}/(\sqrt{3}n)) + 3n \frac{\partial}{\partial n} h(\sqrt{2}/(\sqrt{3}n)), \quad (25)$$

where the second term on the r.h.s is calculated using  $\frac{d}{dx} J_0(x) = -J_1(x)$  as

$$\begin{aligned} \frac{\partial}{\partial n} h(\sqrt{2}/(\sqrt{3}n)) &= \frac{\phi}{4\pi} \sqrt{\frac{2}{\sqrt{3}}} n^{-3/2} \\ &\times \int k^2 dk J_1(k\sqrt{2}/(\sqrt{3}n)) \\ &\times \frac{c(\lambda k)}{(\lambda k)^2 + c(\lambda k)(1 + 4\pi\chi)}. \end{aligned} \quad (26)$$

In Fig. 2, the vortex density  $n$  is depicted as a function of the magnetic field  $H$ . As seen in Fig. 2, the vortex density drastically increases in the small region of the magnetic field around  $H_{c1}^0$  if  $\chi \neq 0$ . For the  $\chi = 0$  case,  $H_{c1}^0$  corresponds to  $H_{c1}$ . For the  $\chi = 0.1$  and  $\chi = 0.3$  cases, the vortices appear below  $H_{c1}^0$ , thus,  $H_{c1}$  is lower than  $H_{c1}^0$ ; actually,  $n$  is a double-valued function of  $H$  in the small region  $H < H_{c1}^0$ . This behavior occurs due to the fact that  $h(r)$  becomes negative at some values of  $r$  as seen in Fig. 1.

### III. MAGNETIZATION CURVE

Using the vortex density  $n$  and the formula

$$B = n\phi = H + 4\pi M, \quad (27)$$

we calculate the magnetization  $4\pi M$  as the function of the applied magnetic field  $H$ . To compare our theory with the

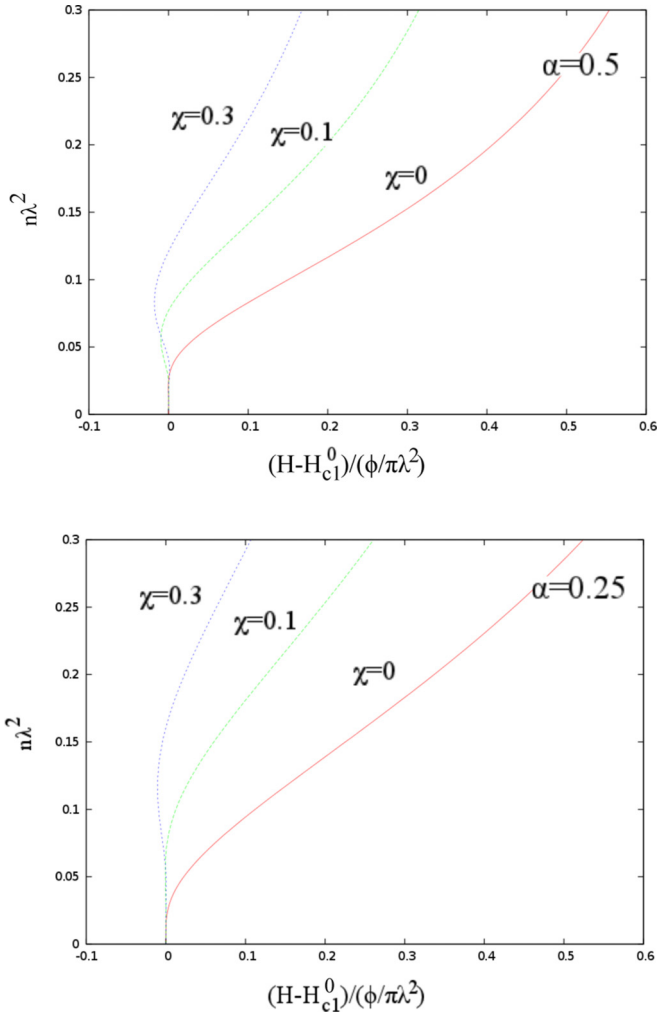


FIG. 2. (Color online) Plots of  $H - H_{c1}^0$  vs  $n\lambda^2$  for two different values of  $\alpha$ . The units of  $H - H_{c1}^0$  is  $\phi/\pi\lambda^2$ . Values of  $\chi$  are 0.3, 0.1, 0.0 from the top to the bottom.

experiment by Das *et al.* [6], we take  $H_{c1}^0 = 5$  Oe from the  $H_{c1}$  value observed in the experiment.  $\lambda$  is taken from the value obtained in the earlier experiment,  $\lambda = 7 \times 10^{-5}$  cm ( $\xi_{ab} = 14$  nm,  $\kappa = 50$  [2]); the volume fraction of the superconducting domains is estimated to be  $0.025/5 = 5 \times 10^{-3}$  using the value of  $4\pi M = -0.025$  gauss at  $H_{c1} = 5$  Oe observed in the experiment [6]. The magnetization curve is shown in Fig. 3. The magnetization increases in the small region of magnetic field around  $H_{c1}^0$ . This behavior is the same as that of the magnetization curve observed in the experiments with use of the superconducting single crystal of  $\text{Cu}_{0.15}\text{Bi}_2\text{Se}_3$  [6]. Since our theory assumes that the vortex density is small and only takes into account the vortex-vortex interaction of the nearest neighbors, it is only adequate in the vicinity of  $H_{c1}$ . The experimental result of Das *et al.* [6] near  $H_{c1}$  resembles the  $\chi = 0$  case.

#### IV. CONCLUDING REMARKS

Our result indicates that the magnetization from the supercurrent carriers is small, suggesting that  $\text{Cu}_{0.15}\text{Bi}_2\text{Se}_3$

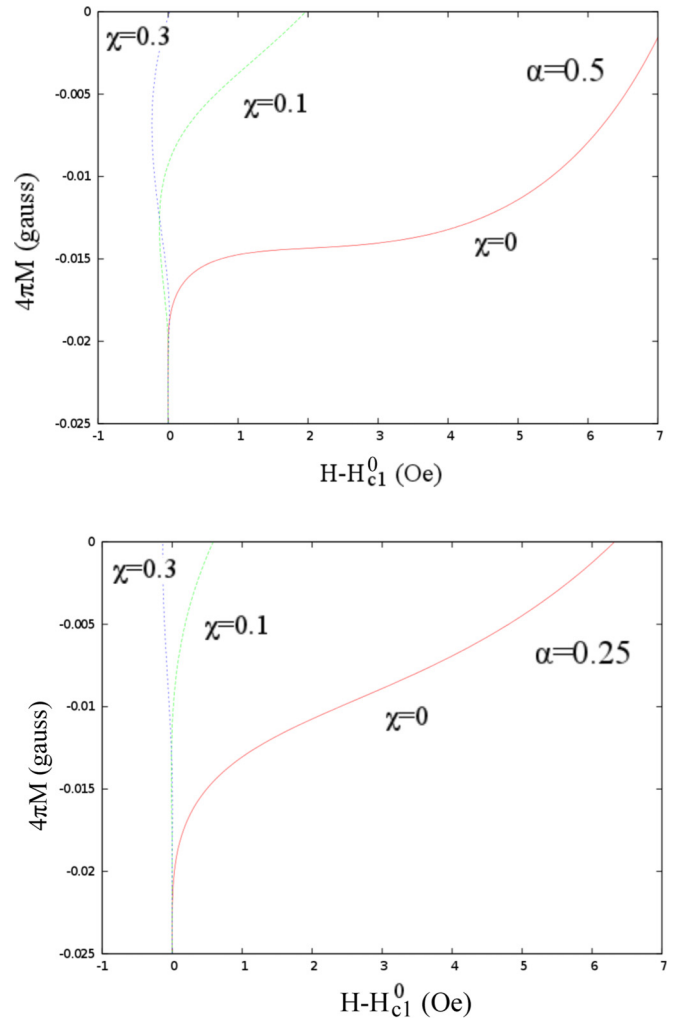


FIG. 3. (Color online) Plots of  $H - H_{c1}^0$  (Oe) vs  $4\pi M$  (gauss) for two values of  $\alpha$  ( $\alpha = 0.25, 0.5$ ). Only parts of the curves where  $n$  is not zero are shown. When  $n$  is zero, the magnetization curve follows  $4\pi M = -HV_f$  due to the Meissner effect, where  $V_f = 5 \times 10^{-3}$  is the volume fraction of the superconducting domains. Values of  $\chi$  are 0.3, 0.1, 0.0 from the top to the bottom.

is not a spin-triplet superconductor. But the possibility of a small value of  $\chi$  cannot be excluded. If  $\chi \neq 0$  is satisfied, the double-valuedness of  $M$  as a function of  $H$  arises around  $H_{c1}$ . This double-valuedness may be observed as a hysteresis of  $M$  depending on increasing or decreasing of  $H$ ; when  $H$  is decreased from the vortex state, the vortices may persist below the  $H_{c1}$  value of increasing  $H$  due to the attractive interaction between vortices.

Taking into account the STM measurement that  $\text{Cu}_{0.15}\text{Bi}_2\text{Se}_3$  exhibits the  $s$ -wave pairing type energy gap [5], one may think that the small  $\chi$  value is the indication of the  $s$ -wave pairing BCS superconductor. However, the doping dependence of the superconducting phase resembles that of the cuprate superconductivity [2]; thus, it is suggested that the recently proposed superconductivity theory for the cuprate based on the spin-twisting itinerant motion of electrons may be relevant [7–11]. The spin-twisting itinerant motion of electrons is stabilized by the strong Rashba spin-orbit interaction in this

system [11], and the doped coppers may become the stabilizing centers of the spin-vortices produced by the itinerant electrons.

More specifically, the supercurrent density given by

$$\begin{aligned} \mathbf{j} &= -\frac{2e}{\hbar} \sum_{\ell}^{N_{\text{loop}}} \lambda_{\ell} \oint_{C_{\ell}} \nabla \psi \cdot d\mathbf{r} \\ &= -\frac{2e}{\hbar} \sum_{\ell}^{N_{\text{loop}}} \lambda_{\ell} \int_{S_{\ell}} \nabla \times (\nabla \psi) \cdot d\mathbf{S} \end{aligned} \quad (28)$$

may be produced upon the copper doping, where  $N_{\text{loop}}$  is the total number of independent loops in the system,  $C_{\ell}$ 's are the independent loops, and  $S_{\ell}$  is the surface with perimeter  $C_{\ell}$ ,  $\psi$  is an angular variable with period  $2\pi$ , and  $\lambda_{\ell}$ 's are material dependent parameters [9–11]. It can be shown that the system

with the above current density exhibits the flux quantum  $hc/2e$  and Josephson frequency  $2eV/h$  ( $V$  is the voltage across the Josephson junction) [7,11].

In this mechanism, the superconductivity is connected to the appearance of points  $\nabla \times (\nabla \psi) \neq 0$ , and positions of the doped coppers become such points. This current flows even in the band insulator situation where an energy gap exists in the single-electron excitation, thus, it can flow in the bulk of the topological insulator. Then, the occurrence of superconductivity is connected to the formation of the spin-vortices produced by the itinerant electrons with spin-twisting motion, in stead of the Cooper pair formation. Since spin vortices can produce small magnetization which is absent in the  $s$ -wave pairing, the hysteresis in the magnetization around  $H_{c1}$  may be observed. The observation (or nonobservation) of it will narrow down the mechanism of the superconductivity in  $\text{Cu}_x\text{Bi}_2\text{Se}_3$ .

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