

Broadening of superconducting resistive transition in macroscopically inhomogeneous Ga–Sb and Zn–Sb alloys under magnetic field

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The effect of magnetic field on superconducting properties of high-resistance metastable Ga–Sb and Zn–Sb alloys, in which the metal–insulator transition takes place, has been studied.

We have detected considerable broadening of the resistive superconducting transition and found that the transition is broader at a higher content of the insulating phase in the sample.

Apart from broadening, the magnetic field induces a mutual shift along the temperature axis of transition curves of resistivity and ac-susceptibility. Our interpretation of these effects is based on an assumption that the typical dimension of conductivity fluctuations in the studied samples is notably smaller than the magnetic field penetration depth λ . As a result, dimensions of the critical-field H_{c2} inhomogeneities are of the same order of magnitude, and small drops of superconducting phase, which lead to a decrease in the sample resistance, but do not affect the magnetic susceptibility, are generated. © 1996 American Institute of Physics. [S1063-7761(96)01807-0]

1. INTRODUCTION

Since properties of high-temperature superconductors (HTSC) have been studied extensively in recent years, a lot of attention has been focused on measurements of resistance under magnetic field, which are greatly affected by interaction between mobile Abrikosov vortices and current.^{1,2} Owing to the high critical temperature T_c and relatively low pinning energy U_p in these superconductors at $T < T_c$,³ thermally activated motion of vortices leading to energy dissipation and hence to broadening of the resistive superconducting transition may be possible in magnetic field H . Besides, current–voltage characteristics of HTSC are nonlinear, which indicates that there are different regimes of vortex motion.

The pinning energy in type II superconductors is considerably higher than T_c , therefore vortices are pinned at currents of up to the critical value $J_c = J_c(H, T)$. For this reason, thermally activated depinning may be detected only at a current close to J_c on the highly nonlinear portion of the current–voltage characteristic, when the effective pinning energy U_p is considerably lower and comparable to the temperature.⁴ This is why the magnetic broadening of resistive superconducting transitions due to the interaction between vortices and current had been discussed only in terms of nonpinned vortex motion before the discovery of HTSC.^{5,6} The most suitable materials for studying free vortices were amorphous alloys, and the agreement between theory and experiment was fairly good.⁷

Another class of superconductors in which the transition is usually broadened by magnetic field are granular materials. In the presence of Josephson coupling among superconducting grains, the superconducting transition broadening in a granular superconductor may be caused by energy dissipation due to phase fluctuations of the order parameter. This effect may be considered as an alternative mechanism of

energy dissipation in HTSC⁸ in addition to the interaction between current and mobile magnetic vortices. This assumption was based on experiments^{9–11} which were conducted at various mutual alignments of current density and magnetic field and demonstrated that the macroscopic Lorentz force is not related to the energy dissipation in HTSC in resistive state under magnetic field.

In granular superconductors, the superconducting transition may be also significantly broadened at zero magnetic field because of macroscopic inhomogeneities leading to fluctuations of superconducting parameters. We are considering, however, another case, when the transition broadening due to the sample inhomogeneity is observed only under magnetic field.^{12,13} This broadening taking place in “dirty” superconductors and caused by the spread of the upper critical field H_{c2} may also occur in both conventional and high-temperature superconductors. The zero-field critical temperature $T_c(0)$ in dirty superconductors is independent of the effective electron free path l ,¹⁴ whereas the upper critical field $H_{c2}(T)$ is a function of the mean free path l :

$$H_{c2}(0) \approx \frac{\Phi_0}{2\pi\xi^2} \propto \frac{1}{l}, \quad l \ll \xi, \quad (1)$$

where $\Phi_0 = \pi\hbar c/e$ is the magnetic-flux quantum and $\xi \propto l^{1/2}$ is the coherence length in the dirty limit. At a temperature close to the critical value, a sample should therefore be divided in the magnetic field into normal and superconducting regions, whose volume ratio is a function of temperature. This results in the transition broadening depending on the sample inhomogeneity, and measurements of the magnetic field broadening yield typical dimensions of inhomogeneities and amplitudes of conductivity fluctuations.¹³

The aim of our work was to elucidate the physical causes of the superconducting transition magnetic broadening in metastable Ga–Sb and Zn–Sb alloys, which undergo a structural transition accompanied by a metal–insulator transition

at higher temperatures. Our results have led us to a conclusion that the transition broadening is due to the spread of the upper critical field.

2. EXPERIMENT

We have studied the superconducting transition in samples of high-resistance Ga–Sb and Zn–Sb alloys in their metastable states obtained by gradually annealing respective materials fabricated under high pressure.^{15,16} An important feature of our experiments is that the critical temperature and width of the superconducting transition at $H=0$ were practically constant in the range of studied states, which allowed us to observe the effect of magnetic field on the transition shape as a function of the sample state.

The samples fabricated under high pressure were metallic polycrystals with a resistivity of about $50 \mu\Omega \cdot \text{cm}$. They could be stored in this state at the liquid-nitrogen temperature for an indefinite time. But at temperatures above 150 K, the alloys gradually transform to an amorphous insulating state. We could study the transformation of their low-temperature characteristics by interrupting the annealing and returning the sample to a liquid-nitrogen temperature. We characterized each state by the parameter

$$q = \log(R_n/R_{in}),$$

where R_n/R_{in} is the ratio of normal resistances in the studied and initial states, respectively, measured at a temperature slightly higher than the superconducting transition, $T_n > T_c$. At zero magnetic field, the values of T_c for the samples of Ga–Sb and Zn–Sb were 4.5 K and 6.5 K, respectively, therefore the temperature T_n was selected to be 6 K for Ga–Sb and 8 K for Zn–Sb.

We measured the electric resistance and ac-susceptibility as functions of temperature and magnetic field in a temperature range of 0.4 to 10 K and under a field of up to 8 T. In measurements of the resistance, the sample was clamped between two pairs of sharpened gold wires, which acted as contacts in the four-terminal configuration for resistivity measurements. The resistivity measurements were conducted at a fixed frequency of 170 Hz using a lock-in amplifier. All the curves of resistivity presented in the paper were recorded on the ohmic regime at all temperatures and magnetic fields. The current conducted across the sample was set on the linear portion of the current–voltage characteristic. As the sample was transformed due to the gradual annealing, the current at which the resistivity could be measured in the linear mode had to be reduced owing to the rise in the normal resistance.

In order to measure the ac-susceptibility, we placed the sample into one of the two secondary windings of a transformer and fed a current at a frequency of 100 kHz to its primary winding. The differential signal from the secondary windings was detected at such a phase so that to record the real component of the magnetic ac-susceptibility. In testing experiments, we reduced the driving current frequency to 1 kHz to verify that the measured effects were independent of the frequency. The maximum intensity of the ac magnetic field h was about 5 Oe at a current across the primary winding of 50 mA.

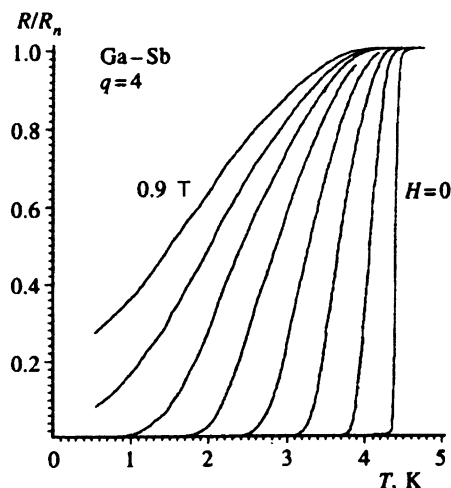


FIG. 1. Effect of magnetic field on the superconducting transition in Ga–Sb.

The results of experiments with both studied materials were similar. Most of the data reported in the paper were measured in the Ga–Sb sample, whose superconducting transition at $H=0$ is more narrow, therefore the effect of magnetic field on its characteristics is more pronounced and more information about its properties can be derived from its measurements.

2.1. Resistive superconducting transition

Superconducting transitions have been detected on the curves of $R(T)$ in various states of the samples in the range of $0 \leq q \leq 6$. This set of the curves combined with records of $R(H)$ at fixed temperatures yielded basic information about the magnetic broadening of the superconducting transition and temperature dependence of the second critical field in samples on various stages of their transformation.

The effect of magnetic field on the superconducting transition in Ga–Sb at $q=4$ is illustrated by Fig. 1, which shows the sample resistance versus temperature normalized to the resistance R_n at $T=6$ K in a temperature range of 0.5 to 5 K at eight equally spaced values of magnetic field ranging between 0 and 0.9 T. These curves indicate that at a field $H \geq 0.5$ T the transition is so wide that the transition temperature cannot be clearly defined. In what follows, we will define the critical temperature T_c as a temperature at which the sample resistance drops by a factor of two, i.e., $R(T_c) = R_n/2$. We will similarly derive the upper critical field H_{c2} from the curves of $R(H)$. Figure 1 demonstrates that the critical temperature defined in this way drops with the magnetic field due to the temperature dependence of the upper critical field H_{c2} .

Let us define the width of the superconducting transition ΔT as a difference between temperatures at which the tangent straight line at the transition midpoint $R = R_n/2$ crosses the horizontal lines $R = R_n$ and $R = 0$. At zero magnetic field the width of the transition is $\Delta T = 0.05$ K and is a flat function of q . Under a magnetic field starting from $H \approx 0.1$ T, the transition is broadened, and its width ΔT increases with both q and H (Fig. 2). Figure 3 demonstrates superconducting transitions recorded at one magnetic field on different stages

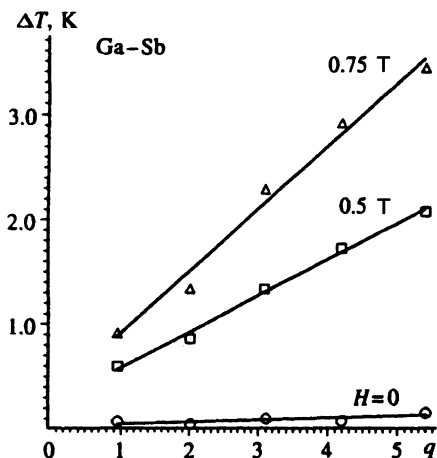


FIG. 2. Width of the superconducting transition at fixed magnetic fields vs stage of transformation in Ga-Sb.

of transformation of the Ga-Sb sample. The transition curve recorded at $H=0$ and $q=3$ is shown for comparison. The set of experimental curves clearly shows that the temperature of the transition onset increases as the sample is annealed, whereas the lower boundary of the transitional region shifts towards low temperatures. At the same time, the critical temperature T_c determined at the midpoint of the transition curve is practically constant with q .

2.2 Magnetic ac-susceptibility

Measurements of the ac-susceptibility χ were carried out concurrently with the resistance measurements and yielded additional information about properties of the studied materials. We recorded χ as a function of both temperature and magnetic field, and studied the effect of the amplitude of the ac electromagnetic field h in the transformer primary coil on the diamagnetic response.

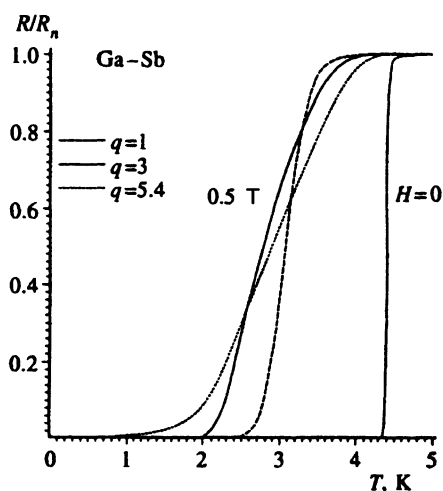


FIG. 3. Superconducting transition in Ga-Sb under a magnetic field of 0.5 T at different stages of transformation. For comparison, the transition at zero magnetic field and $q=3$ is shown.

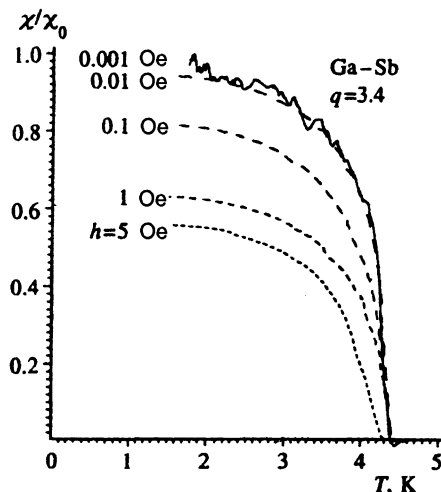


FIG. 4. Magnetic ac-susceptibility normalized to χ_0 in the initial state vs temperature at different amplitudes of the ac field and $H=0$.

In the initial state, the magnetic flux was totally expelled from the sample, and the susceptibility χ_0 was independent of the ac field amplitude. But even on early stages of sample transformation ($q \geq 1$), the diamagnetic response dropped at an ac field amplitude higher than a critical value (Fig. 4), which is typical of granular systems with weakly coupled grains. The lowest curve in Fig. 4 was recorded at the highest electromagnetic field in measuring coils, when the sample heating was quite considerable, which is demonstrated by the 0.1-K shift of the curve towards lower temperatures. Because of the heating, we could not achieve a complete saturation of the diamagnetic response due to the total destruction of weak links in the inhomogeneous sample by the ac field. Nonetheless, the effect of the ac field amplitude on the magnetic susceptibility shown in Fig. 4 is direct evidence that the annealing resulted in weak coupling among sample grains.¹⁷

The most interesting result derived from measurements of ac-susceptibility is that in the magnetic field the transition temperature derived from the curves of susceptibility is lower than that derived from the resistance curves. This effect is shown by the curves in Fig. 5 recorded in the Zn-Sb sample, in which the transition is fairly wide even at $H=0$. The curves $\chi(T)$ are compared to the records of $R(T)$. At zero magnetic field, the temperatures of the transition onset on both $\chi(T)$ and $R(T)$ curves coincide. But under magnetic field the transition onset on the curve of $\chi(T)$ shifts with respect to that on the $R(T)$ curve by the transition width, and under a field $H \geq 0.2$ T it coincides with the point at which the transition on the $R(T)$ curve completes. A similar effect can be seen on the curves of $R(H)$ and $\chi(H)$ for the Ga-Sb sample given in Fig. 6: the sample resistance due to the destruction of superconductivity by magnetic field is detected only when the susceptibility signal vanishes. A similar shift of the transition on $\chi(T)$ curves with respect to that on resistance curves was previously detected in fine-grained superconducting materials¹⁸ in which grain dimensions were notably smaller than the field penetration depth λ . The $\chi(T)$ curves shown in Fig. 5 by solid lines were recorded at a low ac field amplitude. The effect of the ac field amplitude

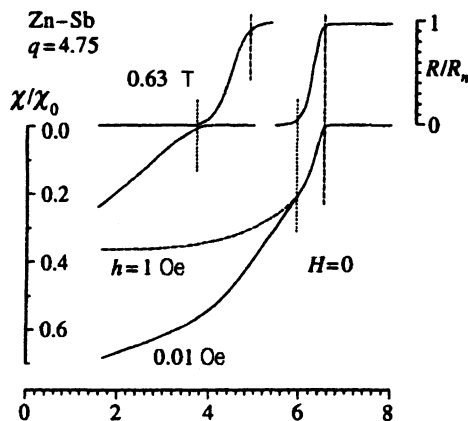


FIG. 5. Effect of magnetic field on the position of superconducting transition detected on curves of resistance vs temperature and of ac-susceptibility vs temperature in Ga-Sb.

on the ac-susceptibility signal in the annealed Zn-Sb sample is similar to that in the Ga-Sb sample (Fig. 4). The drop in the susceptibility signal with the ac field amplitude in Zn-Sb is illustrated in Fig. 5 by the dashed curve.

Figures 5 and 6 indicate that the ac-susceptibility χ drops with the magnetic field. After a long-time annealing of Ga-Sb samples ($q \geq 4$), the signal drops by almost an order of magnitude under a magnetic field as low as $H \approx 0.1$ T. In samples with lower q the ac-susceptibility χ drops in two steps (Fig. 6): first χ drops sharply at a low field, then after a relatively flat portion of the curve, the signal drops to zero at a field of ≥ 0.5 T. At a higher amplitude of the ac field, the function $\chi(H)$ becomes monotonic as in strongly annealed samples. The two-step drop of the diamagnetic response under magnetic field, which was previously observed in granular samples, is due to the destruction of weak coupling by the field and the resulting penetration of magnetic field into superconducting grains.¹⁹

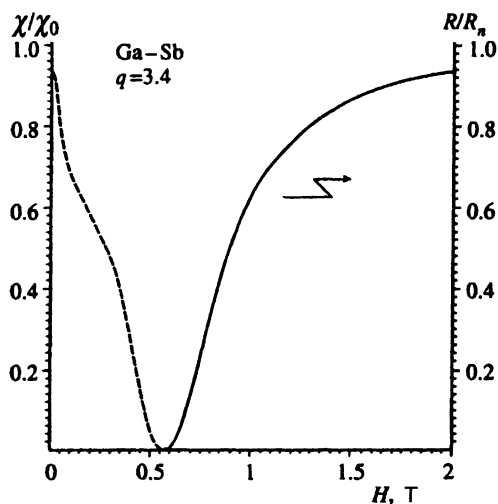


FIG. 6. Signal of ac-susceptibility (dashed line) and resistance (solid line) of the Ga-Sb sample vs magnetic field at $T=1.3$ K and $h=0.01$ Oe.

3. DISCUSSION

The Ga-Sb and Zn-Sb alloys have the following feature: after the transformation of the metastable phase, the sample resistance in the normal state increases by five to six orders of magnitude to hundreds of kilohms, and the resistance as a function of temperature retains its positive derivative, which is typical of metals. Moreover, after a further increase in the resistance, although the derivative dR/dT becomes negative, the extrapolation of the conductivity $\sigma(T)$ to $T=0$ yields a positive value $\sigma(0)$, i.e., from the formal viewpoint, the alloy does not transform to an insulating state even at $q \approx 8$. In order to account for such a property, a model describing the transformation of the dielectric phase as growth of a branching fractal structure was proposed, so it results in considerable lengthening and thinning of current channels.²⁰ Only at $q > 8$ the current channels are intersected by insulating spacers and the resistance as a function of temperature becomes anomalous, i.e., instead of a drop below T_c , a sharp increase is observed.²¹ In this paper we discuss measurements on samples with $q < 5$ and metallic conductivity.

Although our samples have relatively low T_c , the detected effect of magnetic field on the superconducting transition is apparently similar to that observed in high- T_c superconductors.²²⁻²⁴ Moreover, we can select a range of magnetic field in which the resistance around the superconducting transition is fairly well described by an exponent with an activation energy ranging between 10 and 100 K and dropping with the magnetic field. If we assume that the transition broadening in magnetic field is due to the thermally activated motion of vortices, this leads us to a conclusion that typical pinning energies in our samples should be about an order of magnitude lower than in high- T_c samples and two orders of magnitude lower than in conventional superconductors. We do not see any physical reason for such a low pinning energy, especially accounting for the large number of point defects and internal interfaces, which should act as pinning sites.

Our interpretation proposed in this paper is based on the sample inhomogeneity. First of all, note that we deal with dirty superconductors, in which the upper critical field H_{c2} depends on the electron free path, according to Eq. (1). The transformation of our samples to high-resistance states obviously leads to a higher electron scattering frequency owing to at least three processes: (a) the increase in the defect concentration when some atoms are displaced from their equilibrium positions as the metastable phase lattice is destabilized; (b) additional scattering on interfaces between crystalline and amorphous phases generated during the annealing; (c) generation of additional strain fields in conducting channels due to a larger specific volume of amorphous phase (in our samples, this volume increased up to 25% during the annealing¹⁵). It is obvious that these processes not only increase the scattering frequency, but also generate inhomogeneities whose typical dimensions are determined by those of the fractal structure of the insulating amorphous phase.

On the earlier stages of transformation ($q \leq 5$), the increase in the scattering frequency affects neither T_c nor the

shape of the superconducting transition at $H=0$ (Figs. 2 and 3), but significantly changes the upper critical field H_{c2} by virtue of Eq. (1). The superconducting transition broadening is due to the inhomogeneity of the electron free path l , hence of H_{c2} , over the sample volume. The proposed mechanism accounts for the broadening towards higher temperatures, i.e., the local increase in the critical parameters. But one can see in Fig. 2 that the annealing also leads to the transition broadening towards lower temperatures, i.e., regions with lower critical superconducting parameters are generated in the sample. These may be confinements in conducting channels or insulating walls generated in branching of the insulating fractal structure. At zero magnetic field, although the emerging weak links reduce the critical current, they do not affect the overall phase coherence. Therefore the sample may be considered as a Josephson medium discussed in our recent publication.²⁵ Under magnetic field, Josephson coupling is destroyed, which results in the low-temperature "tail" of the transition indicating the existence of regions with lower critical parameters.

Zwicknagl and Wilkins¹³ calculated the effect of magnetic field on the superconducting transition width assuming Gaussian fluctuations of conductivity in space. As a result, they obtained the critical fields H_{c2}^{\max} , H_{c2} , and H_{c2}^{\min} at which the sample resistance equals 90% and 50% of R_n , and zero, respectively, as functions of temperature. In their calculation, the superconducting transition width is controlled by the amplitude of conductivity fluctuations $\delta\sigma/\sigma_0$ and the typical inhomogeneity dimension l_{inh} divided by the coherence length $\xi(0)$, i.e., $l_{\text{inh}}/\xi(0)$. In the strong field limit, i.e., around $T=0$, the parameter H_{c2}^{\max} is explicitly expressed as a function of $\delta\sigma/\sigma_0$:

$$H_{c2}^{\max}(T)/H_{c2}^{\text{BCS}}(T) = 1 + 2\delta\sigma/\sigma_0. \quad (2)$$

Here $H_{c2}^{\text{BCS}}(T)$ is the function $H_{c2}(T)$ in the dirty superconductor limit of the BCS theory for a homogeneous sample described by the Maki-de Gennes equation.²⁶ In the range of intermediate fields,¹³ the maximum critical field H_{c2}^{\max} as a function of temperature is described by the equation

$$1 - \frac{H_{c2}^{\text{BCS}}(T)}{H_{c2}^{\max}(T)} = 2 \frac{\delta\sigma}{\sigma_0} \left[1 - \frac{3}{2} \left(\frac{\xi(0)}{l_{\text{inh}}} \right)^2 \frac{H_{c2}(0)}{H_{c2}^{\max}(T)} \right]. \quad (3)$$

Figure 7 shows critical magnetic fields versus temperature derived from measurements of $R(T)$ and $R(H)$ in Ga-Sb at $q=2$. The experimental curve of $H_{c2}(T)$ is well described by the calculated function $H_{c2}^{\text{BCS}}(T)$ for a homogeneous system with parameters T_c and $(-dH_{c2}/dT)_{T_c}$ of the studied sample. Only in the range of low temperatures the experimental value of H_{c2} is slightly higher than the calculation. A similar deviation was previously detected in inhomogeneous superconductors and discussed by Zwicknagl and Wilkins.¹³ The cause is that experiment yields the mean value \bar{H}_{c2} , which is proportional to $1/\sigma$. Since $H_{c2} \propto 1/\sigma$ and $1/\sigma > 1/\bar{\sigma}$, the measurement of \bar{H}_{c2} is higher than the calculation. From the ratio $H_{c2}^{\max}/H_{c2}^{\text{BCS}}$ in the low-temperature range, we derive from Eq. (2) the value $\delta\sigma/\sigma_0 \approx 0.2$ for the amplitude of conductivity fluctuations.

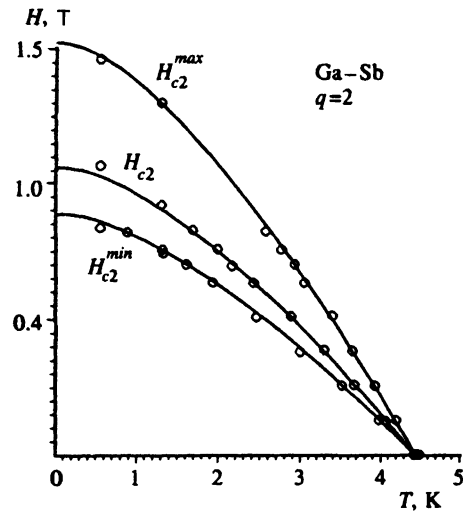


FIG. 7. Critical magnetic fields of the Ga-Sb sample vs temperature.

It follows from the calculation¹³ that the curves of $H_{c2}^{\max}(T)$ and $H_{c2}^{\text{BCS}}(T)$ should converge with the abscissa as the temperature approaches T_c at some point $T_1 < T_c$ controlled by the parameter $\xi(0)/l_{\text{inh}}$ [the expression in square brackets on the right-hand side of Eq. (3) must be zero in this case]. Figure 7, however, indicates that the experimental curves for the three critical fields intersect around $T_c(0)$. The solid curves fitted to the experimental data on $H_{c2}^{\max}(T)$ and $H_{c2}^{\min}(T)$ are calculations by the formula for $H_{c2}^{\text{BCS}}(T)$ with the same T_c , but with different $(-dH_{c2}/dT)_{T_c}$ corresponding to different slopes of the experimental curves at $T_c(0)$. Therefore, with due account of the experimental error owing to the transition width at $H=0$, we can assert that if the experimental curves converge at a finite magnetic field H , this field should be $H \leq 0.1H_{c2}(0)$. Hence follows a lower estimate for the typical inhomogeneity dimension l_{inh} :

$$l_{\text{inh}} = \left(\frac{3}{2} \xi^2(0) \frac{H_{c2}(0)}{H_{c2}^{\max}(T_1)} \right)^{1/2} \geq 4\xi(0) \approx 800 \text{ \AA}.$$

The parameter $\xi(0) \approx 200 \text{ \AA}$ was calculated using Eq. (1) with $H_{c2}(0) \approx 1.1 \text{ T}$ derived from our data on Ga-Sb (Fig. 7).

The upper estimate of the inhomogeneity dimension l_{inh} can be derived from the shift of the superconducting transition on the curve of the ac-susceptibility with respect to that on the resistance curve (Figs. 5 and 6). In order to account for this effect, we only have to assume that the parameter l_{inh} in our samples is smaller than the field penetration depth λ in the superconductor. Then in the temperature range slightly lower than the transition onset, when the sample has a finite resistance, the superconducting inclusions in the sample in the regions where the critical field H_{c2} is higher have small dimensions $d \approx l_{\text{inh}} < \lambda$. Although the total volume of the superconducting inclusions is sufficient to considerably reduce the sample resistance, their contribution to the field screening at $d < \lambda$ is proportional to the factor $(d/\lambda)^2 < 1$. Even at $d \approx \lambda$ the signal due to χ/χ_0 is within 3% to 4%,²⁷ therefore a notable diamagnetic signal at $d < \lambda$

can be detected only after the emergence of an infinite superconducting cluster conducting screening supercurrents along large contours. A numerical estimate of λ can be derived from the curve of $\chi(H)$ assuming that magnetic field can penetrate into a granular superconductor with Josephson coupling at $H \approx H_{c1}$. This estimate for the metastable superconducting Cd–Sb alloy²⁸ yields $\lambda(0) \cong 1200 \text{ \AA}$. It is applicable to both Ga–Sb and Zn–Sb alloys because typical magnetic field in them are close to those in Cd–Sb.

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