Extrinsic paramagnetic Meissner effect in multiphase indium-tin alloys

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The authors report the observation of a paramagnetic Meissner effect (PME) in a two-phase In–Sn alloy in which the phases become superconducting at different temperatures. This observation has been tested and confirmed by constructing an artificial In–Sn sample in which one phase was deliberately encapsulated in another. The authors conclude that PME is extrinsic, rather than intrinsic, and thus describe it as an extrinsic paramagnetic Meissner effect (EPME). It is expected to occur in multiple-phase samples where more than one phase is superconducting and where a suitable microstructural phase distribution is developed. In such samples EPME can be produced at will. © 2006 American Institute of Physics. [DOI: 10.1063/1.2352805]

The reduction to zero of the electrical resistivity in superconducting materials below their critical temperatures (T_c) is a well-documented phenomenon. Concomitantly, the materials become perfect diamagnets for small fields. This effect, termed the Meissner effect (ME), allows for the direct measurement of the transition temperature by superconducting quantum interference device (SQUID) magnetometry. A paramagnetic Meissner effect (PME), i.e., the unexpected observation of a positive magnetic moment in a superconductor below its critical temperature during field cooling (FC), was first reported in 1989 by Svedlindh et al.¹ while studying the high temperature superconductor $Bi_2Sr_2Ca_1Cu_2O_{8+\delta}$. Later, Braunisch *et al.*^{2,3} also reported this "striking" paramagnetism, which they called a PME. The majority of authors who reported PME thereafter in a variety of superconductor materials recognized that some defects, or proximity effects, must be involved in the superconductor sample. Some authors point out that not all measured samples of the same nominal composition material exhibit the PME.⁴ Attempts to develop a more fundamental understanding of this "anti-Meissner" phenomenon range from proposed networks of Josephson junctions⁵⁻⁷ coupled with π junctions,^{8,9} various forms of flux compression,^{4,10-14} and various forms of sample microstructural effects, including structural defects.^{15–19} Interpretations of PME observed in essentially pure materials such as Nb disks,^{14,19} Al disks,²⁰ etc., imply that different portions of a given sample may exhibit subtle differences in the values of T_c , particularly on the surface, and the way in which the magnetic flux is expelled, compressed, or trapped. There remains significant controversy regarding the fundamental origin of the PME.

Here, we report that PME in multiphase In–Sn alloys results from an extrinsic effect: the microstructural encapsulation of grains of one superconducting phase inside grains of a higher T_c phase. This discovery opens the possibility of being able to control the extrinsic paramagnetic Meissner effect (EPME) for potential applications.

We have been investigating the low temperature phase transformations in In–Sn alloys. The phase diagram is shown in Fig. 1.²¹ Although the metallic β -Sn phase is shown to be stable at temperatures above room temperature, in practice it remains metastable also at cryogenic temperatures. Each of the phases, β -Sn, γ -InSn, and β -InSn, becomes superconducting at low temperatures (~3.7, ~4.7, and ~6.2 K, respectively²²) and hence their presence and the amount present can be detected by SQUID magnetometry. Because these phases are stable (or metastable) over a range



FIG. 1. Phase diagram of the system InSn (Ref. 21). The three phases of interest in this investigation are β -Sn (bct, *tI*4), γ -InSn (sh, *hP*1), and β -InSn (bct, *tI*2) with superconducting transition temperatures of 3.7, ~4.2–6.0, and ~5.5–6.5 K, respectively (Ref. 22).

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FIG. 2. (Color online) Temperature dependence of magnetic moment of a $Sn_{90}In_{10}$ alloy by field cooling mode (FC curve) after quench from annealing temperatures of 340 and 380 K, respectively.

of compositions at low temperatures, their superconducting transition temperatures are a function of the particular compositions. In the course of investigating an $In_{10}Sn_{90}$ alloy that contained the β -Sn phase in the γ -InSn matrix we observed the paramagnetic Meissner effect behavior. The particular arrangement of the two phases is the result of a low temperature martensitic or displacive phase transformation (γ -InSn $\rightarrow \beta$ -Sn).²³ This compositionally invariant shear transformation produces a microstructure in which the β -Sn particles are contained within (i.e., encapsulated) in the γ -InSn matrix. Upon cooling through the T_c of the γ -InSn at approximately 4.7 K, the matrix will first exclude and then trap and compress the magnetic flux inside the β -Sn particles, thus inducing the paramagnetic response as shown in Fig. 2. It is important to point out that neither the pure β -Sn phase nor the pure γ -InSn phase shows this effect. It occurs only when there is a mixture of phases. Thus, the observed paramagnetic response in our In-Sn alloy is the result of the specific microstructure, that is, the PME in our alloys is an extrinsic effect in nature, involving two interacting phases rather than intrinsic to a one-phase material.

To test the hypothesis of the extrinsic nature of PME, we constructed a sample with a core of β -Sn, an outside ring of a β -InSn alloy (20 at.% Sn), and an intermediate ring of the γ -InSn phase. The samples were prepared by melting an In₈₀Sn₂₀ specimen around a rod of pure β -Sn under vacuum in quartz tubing. While the β -InSn alloy was molten, a small amount of interdiffusion occurred, such that In diffused into the β -Sn phase, reacting to form a thin ring of the γ -InSn phase between the β -InSn alloy and β -Sn. A schematic cross section of the sample is shown in Fig. 3(a) along with an optical macrograph in Fig. 3(b), including a composition profile [Fig. 3(c)] determined by wavelength dispersive spectroscopy. The composition profile reveals that the β -Sn and β -InSn phases have uniform compositions but the γ -InSn phase exhibits a composition variation with In contents between \sim 75% and 85% Sn as expected from the equilibrium phase diagram. From the composition profile, we would ex-



FIG. 3. (Color online) (a) Schematic of the phase distribution across the manufactured sample, (b) optical macrograph revealing the interfaces between the three phases, and (c) composition profile clearly showing the three distinct phases.

pect that the β -Sn and β -InSn phases should exhibit welldefined critical temperatures. However, due to the smoothly changing composition profile within the γ -InSn phase, we may expect that the T_c in this phase will occur over a range of temperatures. For the SQUID measurements, a 4 mm diameter disk with a thickness of ~ 0.7 mm was prepared from a section of the solidified composite alloy rod. The magnetic field was applied perpendicular to the flat faces of the sample.

The field cooling results of our "manufactured" sample are shown in Fig. 4. There are clear breaks in the curve at temperatures (6.2 and 3.8 K) corresponding to the T_c of β -InSn and β -Sn, respectively. The increase in magnetic moment at 4.7 K is less distinct and corresponds to the critical temperature of the compositionally varying γ -InSn phase. At 6.2 K, the outside phase (β -InSn) becomes superconducting and traps the magnetic flux within the sample. Upon cooling, radial compression of flux causes the positive measured value for the magnetic moment of the sample (PME). The next phase (γ -InSn) becomes superconducting at 4.7 K and further compresses the flux in the β -Sn inner disk. Finally, the β -Sn phase core becomes superconducting at 3.8 K and



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excludes the magnetic flux, giving rise to a drop in the magnetic moment.

Thus, our present experiments indicate that PME is an extrinsic phenomenon requiring the presence of different phases with different T_c values and requiring a specific spatial distribution of such phases. Thus, in our samples, the EPME is clearly a microstructural phenomenon; it is caused by the specific geometry of the phases present. Such a feature is expected whenever multiple superconducting phases coexist in an alloy in such a way that the phase with a lower transition temperature is surrounded by a phase, or phases, with a higher transition temperature(s).

It is possible that the EPME may find some useful cryogenic applications. In order to obtain a dispersed phase in a (parent) matrix of another, a solid-state diffusional precipitation process or an induced martensitic transformation provides the desired mechanism. Both phases should be superconductors, the matrix having higher T_c than the product. Binary or ternary systems that involve the well-studied superconducting phase with the A15 structure and another superconducting structure, or systems similar to In–Sn, are possible candidates. The understanding of how EPME arises in multiphase samples should allow the production of EPME at will rather than as an artifact of sample preparation.

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- Grütz, V. Kataev, S. Neuhausen, B. Roden, D. Khomskii, D. Wohlleben, J. Bock, and E. Preisler, Phys. Rev. B 48, 4030 (1993).
- ⁴G. S. Okram, D. T. Adroja, B. D. Padalia, O. Prakash, and P. A. J. de Groot, J. Phys.: Condens. Matter **9**, L525 (1997).
- ⁵M. Sigrist and T. M. Rice, J. Phys. Soc. Jpn. **61**, 4283 (1992).
- ⁶R. T. Dias, P. Pureur, P. Rodrigues, Jr., and X. Obradors, Phys. Rev. B **70**, 224519 (2004).
- ⁷A. P. Nielsen, A. B. Cawthorne, P. Barbara, F. C. Wellstood, C. J. Lobb, R.
- S. Newrock, and M. G. Forrester, Phys. Rev. B **62**, 14380 (2000). ⁸D. Dominguez, E. A. Jagla, and C. A. Balseiro, Phys. Rev. Lett. **72**, 2773
- (1994).
- ⁹J. Magnusson, M. Björnander, L. Pust, P. Svedlindh, P. Nordblad, and T. Lundström, Phys. Rev. B **52**, 7675 (1995).
- ¹⁰E. Koshelev and A. I. Larkin, Phys. Rev. B **52**, 13559 (1995).
- ¹¹V. V. Moshchalkov, X. G. Qui, and V. Bruyndoncz, Phys. Rev. B 55, 11793 (1997).
- ¹²B.-H. Zhu, S.-P. Zhou, G.-Q. Zha, and K. Yang, Phys. Lett. A **338**, 420 (2005).
- ¹³L. Pust, L. E. Wenger, and M. R. Koblischka, Phys. Rev. B 58, 14191 (1998).
- ¹⁴P. Kostic, B. Veal, A. P. Paulikas, U. Welp, V. R. Todt, C. Gu, U. Geiser, J. M. Williams, K. D. Carlson, and R. A. Klemm, Phys. Rev. B **53**, 791 (1996).
- ¹⁵S. Riedling, G. Bräuchle, R. Lucht, K. Röhberg, H. v. Löhneysen, H. Claus, A. Erb, and G. Müller-Vogt, Phys. Rev. B **49**, 13283 (1994).
- ¹⁶E. Khalil, Phys. Rev. B **55**, 6625 (1997).
- ¹⁷W. A. Ortiz, P. N. Lisboa-Filho, W. A. C. Passos, and F. M. Araujo-Moreira, Physica C **361**, 267 (2001).
- ¹⁸C. Heinzel, T. Theilig, and P. Ziemann, Phys. Rev. B 48, 3445 (1993).
- ¹⁹P. S. Fodor and L. E. Wenger, Physica C **341–348**, 2043 (2000).
- ²⁰A. K. Geim, S. V. Dubonos, J. G. S. Lok, M. Henini, and J. C. Maan, Nature (London) **396**, 144 (1998).
- ²¹T. B. Massalski, H. Okamoto, P. R. Subraminian, and L. Kacprzak, *Binary Alloy Phase Diagrams*, 2nd ed. (American Society for Metals, Materials Park, OH, 1990), Vol. 3, p. 2295.
- ²²M. F. Merriam and M. Von Herzen, Phys. Rev. **131**, 637 (1963).
- ²³S. Chu, C. Yanar, A. J. Schwartz, T. B. Massalski, and D. L. Laughlin, in *Proceedings of the International Conference on Solid-Solid Phase Transformations in Inorganic Materials, Phoenix, 2005*, edited by J. M. Howe, D. E. Laughlin, J. K. Lee U. Dahmen, and W. A. Soffa (The Minerals, Metals & Materials Society, Warrendale, 2005), Vol. 1, p. 529.

¹P. Svedlindh, K. Niskanen, P. Norling, P. Nordblad, L. Lundgren, B. Lönnberg, and T. Lundström, Physica C **162–164**, 1365 (1989).

²W. Braunisch, N. Knauf, V. Kataev, S. Neuhausen, A. Grütz, A. Kock, B. Roden, D. Khomskii, and D. Wohlleben, Phys. Rev. Lett. 68, 1908 (1992).
³W. Braunisch, N. Knauf, G. Bauer, A. Kock, A. Becker, B. Freitag, A.