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Spontaneous vortex phase in a superconducting weak ferromagnet

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In high-T_c superconducting compounds $R_{1.5}Ce_{0.5}RuSr_2Cu_2O_{10}$ the onset of superconductivity occurs at a temperature T_c much lower than the temperature of the phase transition to weak ferromagnetism, and the diamagnetic response arises at the temperature T_d, which is much lower than T_c. The present paper contains experimental data and a theoretical analysis of this phenomenon. These results collectively show convincingly that between T_c and T_d vortices are present in the sample in equilibrium without an external magnetic field (the spontaneous vortex phase). [S0163-1829(98)52022-X]

Coexistence of ferromagnetism and superconductivity has long attracted attention.[1] This nontrivial phenomenon arises from a simultaneous presence of two competing order parameters, spontaneous magnetic moment $\vec{M}_0$, and the complex superconducting order $\Delta=|\Delta|\exp(i\varphi)$. Usually these two order parameters mutually suppress one another. Nevertheless, the coexistence is possible sometimes, as the theory and the experiment have demonstrated.

There are two possibilities to achieve coexistence of ferromagnetism and superconductivity: either ferromagnetism arises inside the superconducting state, or, inversely, the onset of superconductivity occurs in the ferromagnetic state. In the past both the experiment and the theory dealt with the former case.[2–8] They called these materials *ferromagnetic superconductors*. Recently the experiments have been reported in which coexistence of ferromagnetism and superconductivity was revealed in $R_{1.5}Ce_{0.5}RuSr_2Cu_2O_{10}$.[9] In contrast to previous studies which dealt with superconducting ordering stronger than magnetic ordering, to our knowledge this was the first time where the superconductivity arose in the state with a well developed magnetic order parameter: the ratio of the Nèel temperature T_N to the critical temperature T_c of the superconductivity onset was about 4. We believe it was also the first example where the coexistence of ferromagnetism and superconductivity was observed in high-T_c materials. We call this compound the *superconducting ferromagnet* in order to emphasize the difference between it and ferromagnetic superconductors which refer to ferromagnetism arising in the superconducting state.

Ceramic $R_{1.5}Ce_{0.5}RuSr_2Cu_2O_{10}$ samples were prepared by a solid-state reaction. Their chemistry and structure were described in Ref. 9. The onset of superconductivity at $T_c\sim 33$ K was revealed by the observation of a steep drop of resistivity at zero magnetic field. At the same temperature a weak specific-heat kink has also been observed.[10] Scanning tunneling microscopy and spectroscopy data reveal a superconducting gap all over the sample, confirming that the samples do not consist of separate normal and superconducting regions. The weak ferromagnetism at $T<T_N\sim 130$ K was demonstrated by the dependence of the magnetic moment M on the external magnetic field. But the most remarkable property of the superconducting weak ferromagnet was that the diamagnetism arose at temperatures $T_d\sim 20$ K, which is much lower than T_c. The diamagnetism was revealed as a negative magnetic moment on the virgin magnetization curve obtained after the zero-field-cooling (ZFC) process.[9] We stress that ''superconductivity without the Meissner effect'' exists in the interval $T_d<T<T_c$, which is much broader than the width of the superconducting transition. So this phenomenon may not result from an insufficient sample quality.

There was no proper theoretical interpretation on these observations. In the present paper we present experimental data and a phenomenological model describing magnetic properties of these materials. They unambiguously demonstrate that below T_c the equilibrium mixed state arises even without an external magnetic field (the spontaneous vortex phase).

Let us dwell first on the experimental magnetization curve. In a comparatively narrow field interval about a few hundred Oe the magnetization achieves its saturation value when its further growth becomes much slower and linear. It is natural to suggest that this narrow interval corresponds to the domain structure which is finally suppressed by the applied magnetic field. Then the spontaneous magnetic moment M_0 can be obtained by the extrapolation of the saturation plateau to zero external magnetic field (see an example in the inset in Fig. 1). The moment M_0 is believed to be the saturation moment M_{sat} of the Ru sublattice.[9] The temperature dependence of $M_{sat}=M_0$ is shown in Fig. 1, and T_N is defined as the temperature in which M_{sat} vanishes.

Furthermore, we have revealed that the hysteresis phenomena are much stronger in the superconducting state at $T<T_c$ than in the temperature interval $T_c<T<T_N$. One can see it in Fig. 2 where a few hysteresis loops for different temperatures are shown. A strong enhancement of coercivity in the superconducting state is also demonstrated by the plot of the coercive field H_{coer} as a function of temperature (Fig. 3).

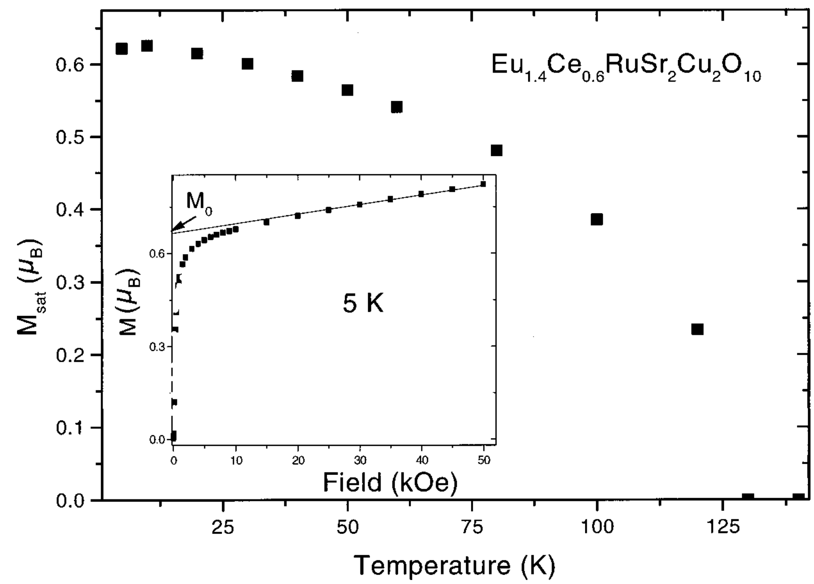

FIG. 1. The dependence of the spontaneous magnetic moment M_0 on temperature. Inset: The spontaneous magnetic moment M_0 is shown for $T=5$ K.

So the most remarkable properties of our superconducting weak ferromagnet are: (i) The diamagnetism arises at temperatures $T_d \sim 20$ K, which is much lower than T_c.[9] (ii) A strong enhancement of coercivity is observed below T_c. Now we shall consider a phenomenological model which explains such a behavior.

The total free energy of our system is

$$F=f(\vec{M}_0)+\frac{1}{2\chi}(\vec{M}-\vec{M}_0)^2+\frac{1}{8\pi}(\vec{B}-4\pi\vec{M})^2 + \frac{\Phi_0^2}{32\pi^3\lambda_0^2}\left(\vec{\nabla}\varphi-\frac{2\pi\vec{A}}{\Phi_0}\right)^2, \tag{1}$$

where χ is the differential magnetic susceptibility, Φ_0 is the magnetic-flux quantum, and λ_0 is the London penetration depth without magnetism. The real penetration depth may be different (see below).

Equation (1) is the London limit of the free-energy expression suggested by Blount and Varma.[2] According to their analysis (see also Refs. 3, 4, 8) interplay between superconductivity and ferromagnetism is governed by electromagnetic effects and the antiferromagnetic components of the order parameter play a negligible role. We also assume that $\vec{M}$ is close to the spontaneous magnetic moment $\vec{M}_0$ at which the free energy $F=f(\vec{M}_0)$ has a minimum in the absence of the superconductivity and the external magnetic field. The minimization of the free energy, Eq. (1), with respect to $\vec{M}$ yields

$$\vec{M}=\vec{M}_0+\frac{\chi}{\mu}(\vec{B}-4\pi\vec{M}_0), \tag{2}$$

where $\mu=1+4\pi\chi$ is the differential magnetic permeability.

In order to find the spatial distribution of the magnetic induction $\vec{B}$, one should look for the minimum of the total free energy with respect to $\vec{A}$. This yields the Maxwell equation for the electric supercurrent $\vec{j}_s$:

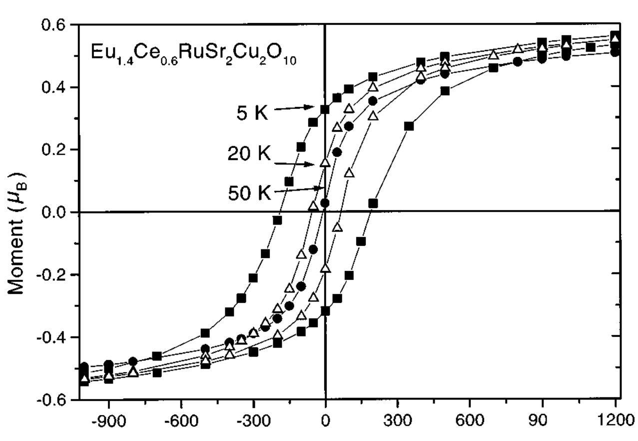

FIG. 2. The hysteresis loops for different temperatures.

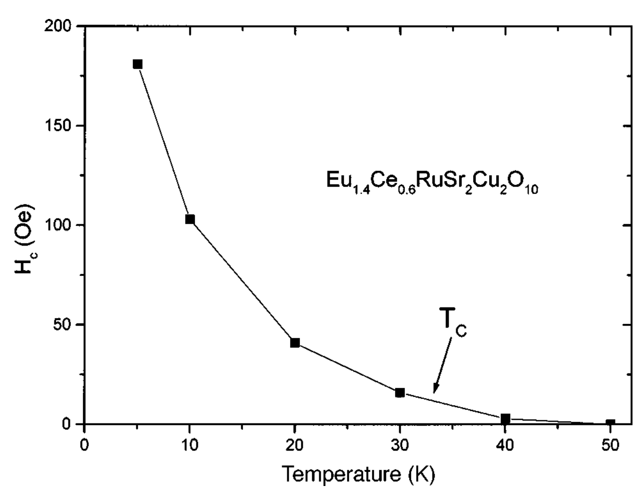

FIG. 3. The coercive field H_{coer} (the width of the hysteresis loop) as a function of temperature.

$$\frac{4\pi}{c}\vec{j}_s=\frac{c\Phi_0^2}{8\pi^2\lambda_0^2}\left(\vec{\nabla}\varphi-\frac{2\pi\vec{A}}{\Phi_0}\right)$$
$$=\frac{1}{\mu}\vec{\nabla}\times(\vec{B}-4\pi\vec{M}_0)=\vec{\nabla}\times\frac{\vec{B}}{\mu}. \quad (3)$$

The Maxwell equation (3) together with the equation

$$\vec{\nabla}\times\vec{j}_s=\frac{c}{4\pi\lambda_0^2}(\vec{B}_v-\vec{B}) \quad (4)$$

yields the London equation which determines $\vec{B}$:

$$\lambda^2\vec{\nabla}\times[\vec{\nabla}\times\vec{B}]+\vec{B}=\vec{B}_v. \quad (5)$$

Here $\lambda=\lambda_0/\sqrt{\mu}$ is the London penetration length into the magnetic superconductor, and $\vec{B}_v$ is the vortex field which takes into account the presence of vortex lines. It is directed along the vortex lines, its magnitude being $B_v=\Phi_0\Sigma\,\delta(\vec{r}-\vec{r}_i)$, where $\vec{r}_i$ is the position vector of the ith vortex line in the plane normal to the line.

Thus the presence of the magnetic moment does not change the distribution of the magnetic induction in the superconductor, except for the renormalization of the London penetration depth.[1] Therefore, in order to find the free energy of the superconducting ferromagnet averaged over the vortex-array cell, we can use the average free energy of the nonmagnetic superconductor:

$$F_0(\overline{B})=\frac{1}{S}\int\left[\frac{B^2}{8\pi}+\frac{\Phi_0^2}{32\pi^3\lambda^2}\left(\vec{\nabla}\varphi-\frac{2\pi\vec{A}}{\Phi_0}\right)^2\right]ds. \quad (6)$$

Here integration is performed over the vortex-array-cell area S in the plane normal to the vortex lines, and $\overline{B}=\Phi_0 n_v$ is the average magnetic induction, where n_v is the vortex-line density. Then the average total energy (1) of the superconducting ferromagnet may be presented as

$$F(B)=f(\vec{M}_0)+\frac{2\pi}{\mu}M_0^2-\frac{1}{\mu}\vec{B}\cdot\vec{M}_0+\frac{1}{\mu}F_0(B), \quad (7)$$

where now and later on $B=\overline{B}$ is the average magnetic induction.

The magnetization curve can be found from the expression for the magnetic field $\vec{H}$:

$$\vec{H}=4\pi\frac{\partial F}{\partial\vec{B}}=\frac{1}{\mu}[\vec{H}_0(\vec{B})-4\pi\vec{M}_0], \quad (8)$$

where $\vec{H}_0(\vec{B})=4\pi\partial F_0/\partial\vec{B}$ is the magnetic field which corresponds to the magnetic induction $\vec{B}$ in the nonmagnetic superconductor. These relations show the transformation by which one can obtain the curve $B(H)$ for the superconducting ferromagnet from the curve $B(H_0)$ for the nonmagnetic superconductor: (i) to shift the curve along the horizontal axis H by the length $4\pi M_0$; (ii) to scale the axis H by the factor $1/\mu$.

The critical fields separating the Meissner and the mixed states are

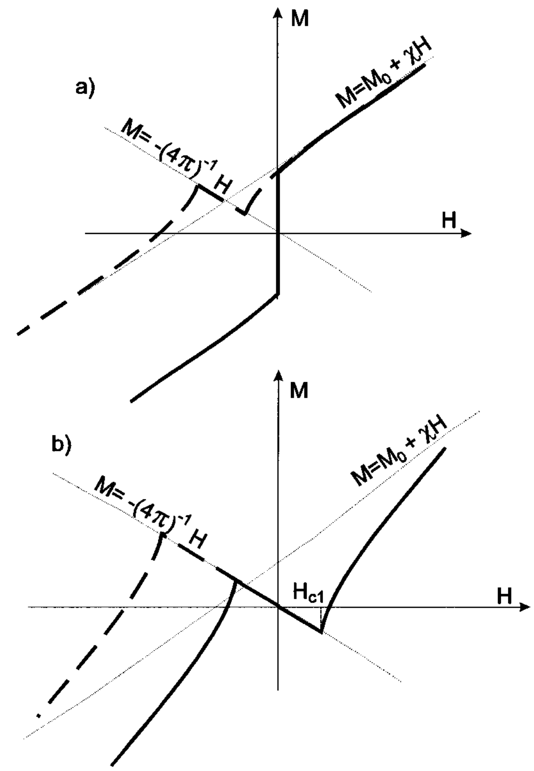

FIG. 4. Schematic theoretical magnetization curves: (a) $T_c>T>T_d$; (b) $T<T_d$. The dashed lines are for the case of fixed $\vec{M}_0$. The thick solid lines show magnetization curves when $\vec{M}_0$ reverses its direction at $H=0$.

$$H_{\pm c1}=4\pi\frac{\partial F}{\partial B}\bigg|_{B\to\pm 0}=\frac{1}{\mu}(-4\pi M_0\pm H_{c1}^0), \quad (9)$$

where H_{c1}^0 is the first critical field for a nonmagnetic superconductor, and signs $+$ and $-$ correspond to the sign of $\vec{B}\cdot\vec{M}_0$ in the limit $\vec{B}\to 0$. Thus for a domain with fixed $\vec{M}_0$ the Meissner state does not vanish, but is shifted and contracted along the axis $\vec{H}$. The magnetization curves for a single-domain sample with fixed $\vec{M}_0$ are shown by dashed lines for $T_d<T<T_c$ in Fig. 4(a) and for $T<T_d$ in Fig. 4(b).

However, the picture given above does not take into account that there are domains with different directions of the spontaneous magnetic moment $\vec{M}_0$. The experiment at $T>T_c$ shows that the external magnetic field remagnetizes the sample very easily, with quite weak hysteresis: $\vec{M}_0$ reverses its direction in a narrow interval of fields. Neglecting this field interval related to the domain structure we have the simplified magnetization curve $\vec{M}=M_0\vec{H}/H+\chi\vec{H}$ at $T>T_c$. Then in order to obtain the magnetization curve in the superconducting state ($T<T_c$), one must take the part of the magnetization curve at $H>0$ obtained above for a single-domain structure with fixed $\vec{M}_0$ and supplement it by its space inversion [solid lines in Figs. 4(a) and 4(b)]. On the magnetization curve obtained by this procedure, the Meissner state is possible only at $T<T_d$ when $4\pi M_0$ becomes smaller than the first critical field H_{c1}^0 of the nonmagnetic superconductor with the free energy $F_0(B)$ [Fig. 4(b)]. Above T_d at zero H the whole sample is in the mixed state,

but there are domains with opposite directions of the magnetic flux corresponding to the directions of the spontaneous moment $\vec{M}_0$.

Until now our model assumed that the critical magnetic field for the Meissner state exceeded the field which suppresses the magnetic-domain structure. In order to make our model even more realistic, one should take into account that in the experiment both fields are of the same order. Since we do not have enough information on the material parameters necessary for the knowledge of the domain structure, we restrict ourselves with some general comments on a possible modification of the magnetization curve in the presence of the domain structure. At $T<T_d$ and at low magnetic fields there is a mixture of domains which are in the Meissner state and of those in the mixed state. This means that one can *never* observe the full Meissner response with the susceptibility $\chi=-1/4\pi$, but it is still possible to observe the negative susceptibility $-1/4\pi<\chi<0$.

We believe that the last case is really observed in our experiment at $T<T_d$; at low magnetic fields the magnetic response comes from different domains which are either in the Meissner, or in the mixed state. Therefore one cannot observe the full Meissner response. Thus our sample is inhomogeneous on two spatial scales: (i) a smaller scale of the order of the intervortex distance in the mixed-state domains, and (ii) a larger scale which is the typical size of the domains themselves. It is necessary to emphasize, however, that this inhomogeneity is induced by the broken symmetry of the ordered state, and has nothing to do with the possibility of the granular structure of the material itself. The latter possibility is quite improbable as was discussed in Ref. 9. An assumed domain structure with a mixed state in some domains must arise even in a sample which is ideally homogeneous above T_N.

Now let us make a rough estimation of the ratio between M_0 and H_{c1}^0, which is crucial for our interpretation. At temperatures of the order or less than T_c the spontaneous magnetic moment M_0 has no essential temperature dependence and is about 34 Oe (see Fig. 1). As a value of H_{c1}^0, one may choose the typical value of the first critical field in the nonmagnetic superconductors of a similar structure: $H_{c1}^0\sim[1-(T/T_c)^2]400$ Oe. We see that at the temperature $T_d\sim 20$ K where the diamagnetic response appears, H_{c1}^0 is of the same order as $4\pi M_0$. This qualitatively confirms our model.

In our theoretical analysis we restricted ourselves with the equilibrium magnetization curve. As was already mentioned the irreversibility (the hysteresis loop) is more pronounced in the superconducting state. This means that irreversibility arises mostly from the vortex pinning, but not from the pinning of domain walls responsible for coercivity in common ferromagnets.

Thus we have shown that in a superconducting ferromagnet at $T_c>T>T_d$ magnetic-flux lines are present in a sample in equilibrium even without an external magnetic field. This state was called the *spontaneous vortex phase*.[4] Recently Ng and Varma[8] suggested looking for the spontaneous vortex phase experimentally in $ErNi_2B_2C$ at temperatures below 2.3 K. One of their papers is titled ''Spontaneous Vortex Phase Discovered?'' We believe that the question mark can be removed; the spontaneous vortex phase has *already* been discovered in the superconducting weak ferromagnets $R_{1.5}Ce_{0.5}RuSr_2Cu_2O_{10}$.

In summary, our observations and the phenomenological model have demonstrated the coexistence of weak ferromagnetism and superconductivity in high-T_c materials under the condition that the magnetic ordering is stronger than the superconducting ordering ($T_c\ll T_N$). In this case the diamagnetic response becomes possible at temperatures $T<T_d$, which is much lower than the temperature T_c. In the temperature interval $T_d<T<T_c$ magnetic-flux lines are present in equilibrium without an external magnetic field (spontaneous vortex phase), but there are domains with opposite directions of the magnetic flux.

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