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Internal magnetic field of superconducting ferromagnets

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We have measured the nonlinear response to the ac magnetic field in the superconducting weak ferromagnet Ru-1222, at different regimes of sample cooling which provides unambiguous evidence of the interplay of the domain structure and the flux distribution in the superconducting state. This is a proof of coexistence of ferromagnetic and superconductive order parameters in high-T_c ruthenocuprates.

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The problem of coexistence of superconductivity (SC) and ferromagnetism (FM) has been studied for almost 50 years starting from the theoretical work by Ginzburg[1] (see also Ref. 2). Coexistence of weak-ferromagnetism (W-FM) and SC was discovered some time ago in $RuSr_2R_{2-x}Ce_xCu_2O_{10}$ (R=Eu and Gd, Ru-1222) layered cuprate systems,[3] and more recently[4] in $RuSr_2GdCu_2O_8$ (Ru-1212). The SC charge carriers originate from the CuO_2 planes and the W-FM is related to the Ru layers. In both systems, the magnetic order does not vanish when SC sets in at T_c, and remains unchanged and coexists with the SC state. The Ru-1222 materials (for R=Eu and Gd) display a magnetic transition at $T_N = 125-180$ K and bulk SC below $T_c = 25-50$ K ($T_N > T_c$) depending on the oxygen concentration and sample preparation. This discovery has launched a new wave of investigations in this field.[5] The problem is of general interest for condensed matter physics and is relevant for many materials, in particular unconventional superconductors (including some heavy fermions) with triplet pairing.[6]

Despite a lot of work done in the past and recently, debates concerning whether such coexistence is genuine are still continuing. Evidence in favor of coexistence is mostly indirect and refers to some peculiarities of the magnetization curve. One of the most pronounced manifestations of SC-FM coexistence is the spontaneous vortex phase (superconducting vortices induced by the internal magnetic field from the FM magnetization). It explains well the magnetization curve of these materials (see Ref. 7 and references therein). However, this phase has not yet been observed experimentally (visualized as the more common mixed state of type-II superconductors).

In the past the evidence of the FM-SC coexistence referred mostly to the magnetic properties of the materials affected by the presence of superconductivity. In this paper we present an additional experimental evidence of the effect of the ferromagnetic order parameter, on the superconducting order parameter. The ferromagnetic order parameter, namely the spontaneous magnetization, is a source of an internal magnetic field inside a sample even without an external magnetic field H. On the other hand, the superconducting properties of type-II superconductors depend strongly on whether the sample was cooled to the SC state in zero magnetic field (ZFC) or in a finite magnetic field (FC). Here a "field" is supposed to be an *external* magnetic field. We show here that these properties depend also on the *internal* magnetic field during the cooling process. We exploited the procedure, which we shall call the internal-field cooling (IFC): The sample was cooled down to T_{IFC} under an external magnetic field H_{IFC} ($T_{IFC} < T_N$). At T_{IFC}, H_{IFC} was turned off and further cool down to $T = 5$ K was done at $H = 0$. It appears that, by using the IFC procedure, the properties of the SC state were different from those measured after the regular ZFC process from temperatures above T_N. Thus, in the SC state, the sample senses the internal magnetic field evolved from the remnant magnetization, which was formed in the normal ferromagnetic phase and then frozen at further cooling.

We measured the nonlinear response to the ac magnetic field, which is a sensitive probe of superconducting vorticity, as demonstrated by numerous investigations in the past.[8–10] Ceramic sample of $Gd_{1.5}Ce_{0.5}Ru_2Sr_2Cu_2O_{10}$ (Ru-1222) with dimensions $8 \times 2 \times 2$ mm^3 was prepared by a solid-state reaction as described in Ref. 3.

In a nonlinear medium, magnetization oscillations, induced by an ac magnetic field $h(t) = h_0 \sin \omega t$, may be expanded in a Fourier series,

$$M(t) = h_0 \sum_{n>0} \chi_n' \sin(n\omega t) - \chi_n'' \cos(n\omega t), \tag{1}$$

where χ_n' and χ_n'' ($n = 1, 2, 3, \ldots$) are the in-phase and out-of-phase components of the harmonic susceptibility. In all experiments described here we measured the voltage drop induced in a pickup coil, which is proportional to the time derivative of $M(t)$. Our homemade experimental setup was adapted to a commercial MPMS SQUID magnetometer. An ac field $h(t)$ at a frequency of $\omega/2\pi = 1.5$ kHz and an amplitude up to the $h_0 = 3$ Oe was generated by a copper solenoid existing inside the SQUID magnetometer. The temperature, dc magnetic field, and amplitude dependencies of the fundamental and third harmonic signals presented here have been measured by the two coils method.[9] In the present paper the results for the direct and third harmonics will be discussed.

Figure 1 shows the temperature dependencies of the in-phase susceptibility χ_1' and of the amplitude of the third harmonic $A_{3\omega} \propto h_0 |\chi_3' - i\chi_3''|$, measured after the ZFC process at $H = 0$. The temperature dependence of χ_1' is typical for superconducting ferromagnets.[3] This plot reveals three transitions, (i) the paramagnetic-antiferromagnetic transition at $T_N \approx 125$ K, (ii) the most pronounced transition, which cor-

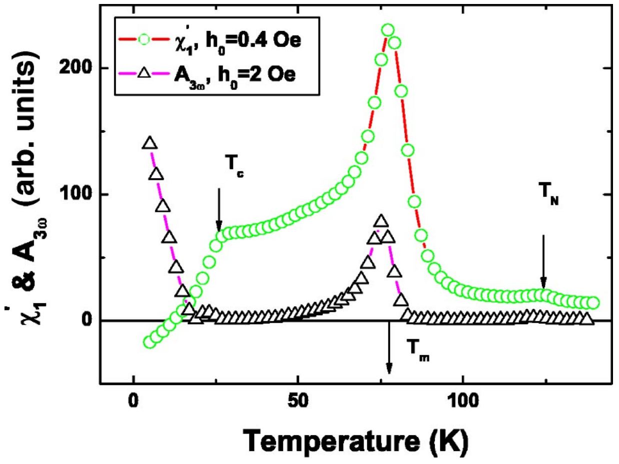

FIG. 1. (Color online) Temperature dependencies of χ_1' and $A_{3\omega}$.

responds to the peak at $T_m \approx 78$ K, and (iii) the transition into the SC state at $T_c \approx 28$ K. The nature of the second transition, which is evident both in the linear and the nonlinear response, is not yet completely clear and is discussed elsewhere.[3,11] Ambiguity is connected with the magnetic phase between T_m and T_N, which is characterized by low coercivity. On the other hand, the $T_c < T < T_m$ temperature region definitely corresponds to the weak-ferromagnetic phase.[3]

The third harmonic behavior is different for $T > T_c$ and $T < T_c$. For $T > T_c$ the behavior is typical for ferromagnetic materials and was known already from Rayleigh's investigation on iron.[12] The third harmonic response demonstrates a quadratic dependence on h_0 (inset in Fig. 2), which directly derived from the oscillatory motion of the domain walls.[13] This signal should decrease at low temperatures and it becomes unobservable under our experimental conditions. For $T < T_c$ the third harmonic grows very fast with temperature decreasing (Fig. 1), and its dependence on the ac field amplitude (Fig. 2) is different from that at $T > T_c$, as evident from the saturation for the nonlinear response at high amplitude of excitation, instead of a quadratic growth. The growth of the nonlinear response in the superconducting materials was revealed in numerous previous experimental investigations.[8–10] Various mechanisms were suggested for this nonlinear response based on the critical state model[8] and the presence of weak links.[10] In particular, the response shown in Fig. 2 is well described by the Josephson-media model. We do not have to discuss these models, since all of them relate the response to the penetration of the magnetic flux (vortices) into the sample, and only this fact is essential for the present investigation. Thus it seems reasonable that the $A_{3\omega}$ at $T < T_c$ is an effective probe of the vortex distribution in the superconducting media.

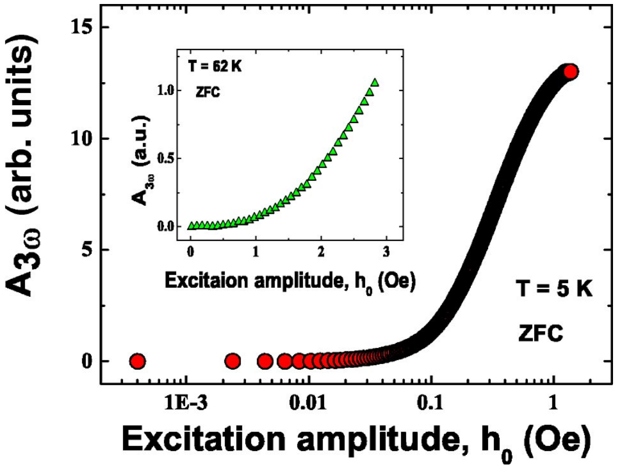

FIG. 2. (Color online) Amplitude dependencies of $A_{3\omega}$ at $T=5$ K. Inset, amplitude dependence of $A_{3\omega}$ in magnetic phase at $T=62$ K.

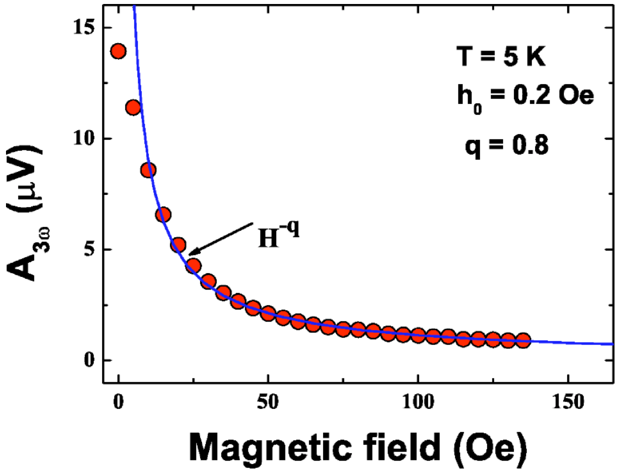

FIG. 3. (Color online) Magnetic field dependence of $A_{3\omega}$ at $T=5$ K after ZFC.

Figure 3 demonstrates ZFC dependence of $A_{3\omega}$ on the external magnetic field. One can see that $A_{3\omega}$ decreases with the magnetic field. At high magnetic fields $A_{3\omega}$ is a power function of the H, $A_{3\omega} \propto H^{-q}$, with $q \approx 0.8$. Suppression of the $A_{3\omega}$ by the magnetic field applied after ZFC was observed in the previous nonlinear studies and agrees with all suggested models of the nonlinear response. The nonlinearity under discussion is connected with a nonhomogeneous distribution of the magnetic flux, which penetrated into the sample, and the magnetic flux distribution becomes more and more uniform, when the vortex density increases. On the other hand, in the Meissner state the nonlinear response must be quite weak, and the magnetic field dependence of $A_{3\omega}$ should have a peak at some H, as was observed in some materials.[8] But in ceramics with numerous weak links, such as our material, this field can be extremely small, and the peak is not observable. Moreover, we deal with the superconducting ferromagnets, where the spontaneous vortex phase can replace the Meissner state at $H=0$. Altogether this explains why we observe the maximum value of $A_{3\omega}$ at $H=0$.

Now let us consider the experimental results in the IFC process. After turning off the magnetic field H_{IFC} at temperature T_{IFC}, the sample was cooled in $H=0$ down to $T=5$ K and the signal of the third harmonic at $T=5$ K was measured. Figure 4 shows $A_{3\omega}(H_{\mathrm{IFC}})$ dependence for $T_{\mathrm{IFC}}=40$ K and 70 K. It is evident that the field H_{IFC} suppresses the $A_{3\omega}$ signal similarly to the external field after ZFC in Fig. 3 even

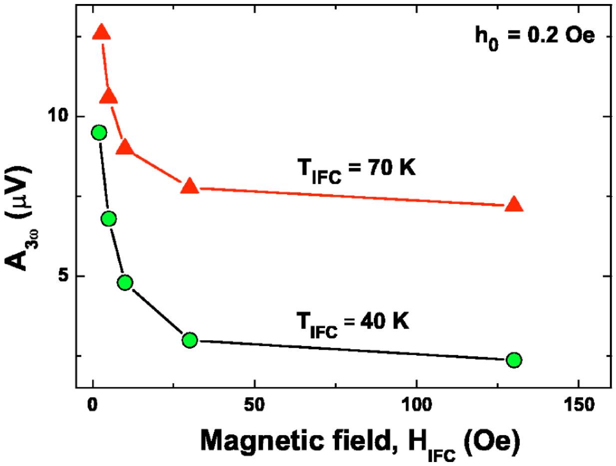

FIG. 4. (Color online) $A_{3\omega}(T=5\text{ K})$ as a function of H_{IFC} for $T_{\mathrm{IFC}}=40$ K and 70 K.

though H_{IFC} was turned off *before* the onset of superconductivity. Turning off H_{IFC} at $T=40$ K affects $A_{3\omega}$ more strongly than for $T=70$ K due to larger remanent magnetization at $T=40$ K. This behavior is typical for the FM materials.[13]

Figure 5 presents the signal of the third harmonic $A_{3\omega}(T=5\text{ K})$ as a function of T_{IFC} after cooling in $H_{\mathrm{IFC}}=30$ Oe. The signal of the $A_{3\omega}(T=5\text{ K})$ decreases for $T_{\mathrm{IFC}}<T_m$. This demonstrates that the suppression of the third harmonic response by the internal magnetic field takes place only if the field cooling continues down to the weakly ferromagnetic phase with essential coercivity. It is known[7] that in idealized single-domain superconducting ferromagnets the internal magnetic field from the spontaneous magnetization $4\pi\vec{M}$ has the same effect on the phase diagram, i.e., on the magnetic flux penetrating into the sample, as the external field. This can be generalized in the more realistic case of a multidomain sample with nonzero average internal field $4\pi\langle\vec{M}\rangle$. On the basis of this argument we can use the plot of $A_{3\omega}(H)$ (Fig. 3) as a calibration curve to estimate the magnitude of the frozen internal magnetic field (H_I). Namely, we take the value of $A_{3\omega}$ from the plot in Fig. 4, find the value of H, which corresponds to this value of $A_{3\omega}$ in Fig. 3, and assume that this value of H gives a reasonable estimation of H_I. Figure 6 presents the dependence of H_I on H_{IFC}.

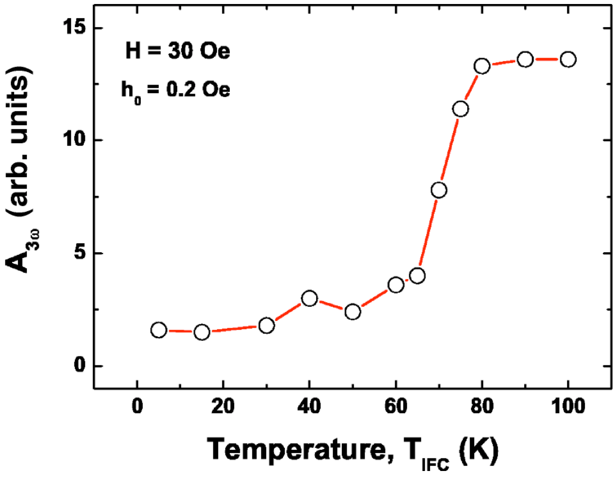

FIG. 5. (Color online) Amplitude of the third harmonic $A_{3\omega}$ at $H=0$ and $T=5$ K vs T_{IFC}.

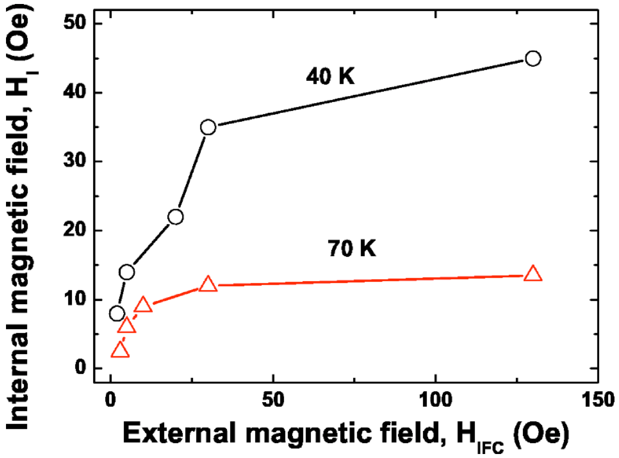

FIG. 6. Internal magnetic field vs H_{IFC} for $T_{\mathrm{IFC}}=40$ K and 70 K.

The internal magnetic field arises from the frozen remanent magnetization $4\pi\langle\vec{M}\rangle$ after field cooling down to T_{IFC}. We have compared Fig. 6 with direct dc remanent magnetization measured in our previous studies.[15] It appears that there is a reasonable agreement (with an accuracy of ±20%) between the two methods, and confirms our scenario.

The phenomenon revealed in our experiment is possible if the domain structure formed in the ferromagnetic phase can be frozen down to the superconducting state. On the other hand, as was noted in the pioneering paper by Ginzburg[1] and confirmed by the detailed analysis in Ref. 14, superconductivity should strongly affect the equilibrium domain structure: Its period should grow, and in equilibrium any sample in the Meissner state is a single domain. But in our case we deal with a nonequilibrium domain structure, which is a metastable state due to coercivity.

The presence of the frozen internal field in the superconducting phase clearly demonstrates that the sample is in the mixed state with many vortices. One cannot call this state the spontaneous vortex phase because the latter refers to the *equilibrium* state, but we deal with a metastable state. We have analyzed here the nonlinear response, which is sensitive to the average internal field $4\pi\langle\vec{M}\rangle$. The absolute value of the average magnetization $\langle\vec{M}\rangle$ is less than the saturation magnetization M, which can determine the vortex density in a single-domain sample.[7] However, the saturation magnetization may create vortices inside domains. Since $\vec{M}$ changes its direction from domain to domain, we obtain the vortex tangle, which does not contribute to the average internal field $\sim 4\pi\langle\vec{M}\rangle$, studied here. This vortex tangle is expected to exist even after the ZFC process and contributes to the initial value of the third harmonic, which was detected without external or internal magnetic field. These arguments illustrate that the magnetic flux distribution in a real (especially ceramic) superconducting ferromagnet can be very complicated. Genuinely zero field cooling is practically impossible: if one cools a sample in zero external field, one cannot avoid internal magnetic fields from the spontaneous magnetization, even if these fields vanish on average but still remain inside the domains. A more detailed analysis of the magnetic-flux distribution would become possible if further investigations provided more information on the structure of the material: sizes of grains and domains, data on crystal anisotropy, etc.

In summary, our measurements of the nonlinear response unambiguously demonstrate the coexistence of the superconducting and ferromagnetic order parameter in Ru-1222 samples below the superconducting critical temperature. Coexistence is manifested by the clear effect on the domain structure frozen from normal FM phase on superconducting properties. We tend to believe that the effect revealed in Ru-1222 is general and can be observed in other materials with FM-SC coexistence.

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