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Effect of ion irradiation on surface morphology and superconductivity of $BaFe_2(As_{1-x}P_x)_2$ films

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ABSTRACT

We have irradiated epitaxial thin films of $BaFe_2(As_{1-x}P_x)_2$ with $x \simeq 0.2$ (optimal doping) with Au ions having an energy of 250 MeV. We have used two different fluences, $\Phi_1 = 2.4 \times 10^{11}$ cm^{-2} and $\Phi_2 = 7.3 \times 10^{11}$ cm^{-2}, and we have studied the effects of irradiation on the surface morphology, on the resistivity and on the critical temperature. We have found that irradiation progressively destroys the very clear and interconnected growth terraces typical of the pristine surface, leading first to their smoothening – accompanied by the appearance of localized defects – and then to a completely disordered surface. The residual resistivity increases by almost 60%, but the critical temperature decreases very little (i.e. by about 2%) on going from the pristine film to the most irradiated one. The possible role of the substrate in these results is discussed.

1. Introduction

Irradiation of superconductors has been widely used for different purposes. The most classic one is the introduction of controlled levels of defects that, in turns, allows controlling quantities of interest in applications, such as the critical current and the irreversibility field. In other cases, irradiation is used to probe the radiation hardness of the materials under study, in view of their applications in space or in particular environments. With the discovery of the iron-based superconductors in 2008, irradiation has also turned out to be a possible tool to discriminate between the two proposed symmetries of the order parameter in these compounds, i.e. s++ (with order parameters of the same sign on all the Fermi surface sheets) and $s\pm$ (with a sign change between holelike and electronlike Fermi surface sheets). Indeed, predictions have been made about the effect of scattering (induced by disorder) on the critical temperature in both these situations.

According to Anderson's theorem [1], in a single-band BCS superconductor with an s-wave order parameter, scattering from non-magnetic impurities should have no effect on either the critical temperature or the energy gap. If the gap is anisotropic but has symmetry-protected node lines, as in the case of d-wave symmetry, scattering averages over different parts of the Fermi surface leading to a gap suppression, to an enhancement of low-energy quasiparticles, and to Andreev bound states. Thus, non-magnetic impurities are pair-breaking in this case. If the gap has nodes that are not symmetry-protected, like in the nodal s-wave symmetry, the averaging mechanism can displace the nodes even removing them and restoring a fully-gapped state [2].

The case of iron-based superconductors is more complicated by the presence of multiple energy bands and by the fact that accidental node lines on the Fermi surface can occur under suitable conditions. Without going too much into details, that can be found elsewhere [3], the effect of disorder (i.e. non magnetic impurities) on T_c depends not only on the gap symmetry ($s\pm$ or s++) but also on the relative amount of *interband* and *intraband* scattering. In particular, for the $s\pm$ symmetry, the maximum rate of T_c suppression (equal to that predicted by Abrikosov–Gor'kov law) is expected for purely interband scattering and when the gaps are equal in amplitude but opposite in sign [3]. Moreover, a transition from $s\pm$ to s++ induced by disorder (with one of the gaps being driven progressively towards the other) has been predicted when the electron–electron coupling constant averaged over the Fermi surface is positive [4]; recently, a disappearance and reappearance of the small gap in $Ba(Fe,Co)_2As_2$ films irradiated with protons has been observed, which would exactly confirm this picture [5].

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It must be kept in mind that all the theoretical predictions deal with *ideal* scattering centers, such as for example Frenkel pairs (vacancy/interstitial) so that some caution must be used in comparing them with experimental results. In particular, the kind of defects produced by irradiation depends on what are the particles in the beam. In Fe-based materials, electrons [2,6,7], protons [8,9], neutrons [10], α particles [11] and heavy ions [12–15] have been used. Electrons, protons and α particles create defects that are mainly point-like, while neutrons also create small cascades of point defects; finally, heavy ions give rise to columnar (or at least correlated) defects along their trajectory, but also spread point-like defects in a bigger volume because of secondary electrons created in the collisions. While the former are fundamental for vortex pinning and thus dramatically affect the irreversibility field and the critical current, the latter are expected to play the leading role in the determination of the transport bulk properties (i.e. resistivity).

In this paper we will describe the effects of Au ions irradiation on thin films of $BaFe_2(As_{1-x}P_x)_2$ around optimal doping. We will show that irradiation dramatically affects the surface morphology and the residual resistivity, while the critical temperature changes very little, decreasing by only 2% for a fluence as high as 7.3×10^{11} cm^{-2}. The comparison of these results with those obtained in single crystals suggest that the substrate of the films may play an important role, possibly overwhelming the effect of the disorder directly induced in the film by irradiation.

2. Thin film growth

The starting $BaFe_2(As_{1-x}P_x)_2$ thin films were grown on MgO single crystal substrates by molecular beam epitaxy, (MBE) with a background pressure of the order of 10^{-7} Pa. All elements were supplied from Knudsen cells using solid sources, as described elsewhere [16,17]. Pure metal sources were used for Ba, Fe, and As, while GaP was chosen as a source of P; an almost pure phosphorus flux was obtained by removing Ga using two trapping caps placed on the crucible. The phosphorous content x of the films was controlled by tuning the P vapor pressure while keeping the vapor pressure of As constant, and the actual stoichiometry of the final films was checked by electron probe micro-analysis (EPMA). TEM images show no reaction layer at the interface between the film and the substrate, and confirm the absence of appreciable defects and grain boundaries [18].

In the following we will refer to two films, “N.316” and “N.360”, that were grown at a temperature of 850 °C and had the same thickness of about 50 nm, but slightly different P content, i.e. $x = 0.19$ and $x = 0.20$, respectively. As shown elsewhere [16], these x values correspond to the top of the superconducting dome in the phase diagram of $BaFe_2(As,P)_2$ films on MgO. Indeed, the critical temperatures of these films are very high (exceeding 30 K).

3. Irradiation with Au ions

The 1.0 cm × 1.0 cm pristine films were cut into rectangular, identical pieces (1.0 cm × 0.25 cm), two of which were homogeneously irradiated with 250 MeV Au ions at the Tandem accelerator of INFN-Laboratori Nazionali di Legnaro [19], with the fluences $\Phi_1 = 2.4 \times 10^{11}$ cm^{-2} and $\Phi_2 = 7.3 \times 10^{11}$ cm^{-2}. During irradiation, the samples were mounted on a copper plate and the particle flux was always kept below 2.5×10^8 s^{-1} cm^{-2} in order to limit the heating of the sample. In iron-based bulk superconductors, in particular Fe(Se,Te) [14] and $Ba(Fe,Co)_2As_2$ [13] single crystals, Au ions of 200–250 MeV were found to produce correlated defects of nanometric cross-section, with a metallic core [14]. Simulations of the energy loss of the ions used in our experiment, carried out with the SRIM-2013 code [20] indicate that: (i) the ions cross the whole film thickness, creating a homogeneous defect distribution into the superconductor, due to the elastic scattering against target nuclei and to ionization phenomena (see Fig. 1a), the second process being more efficient than the first; (ii) the ions lose most of their energy in the substrate (see Fig. 1b) and get implanted in the substrate at a depth of 14.5 μm, as shown in Fig. 1c.

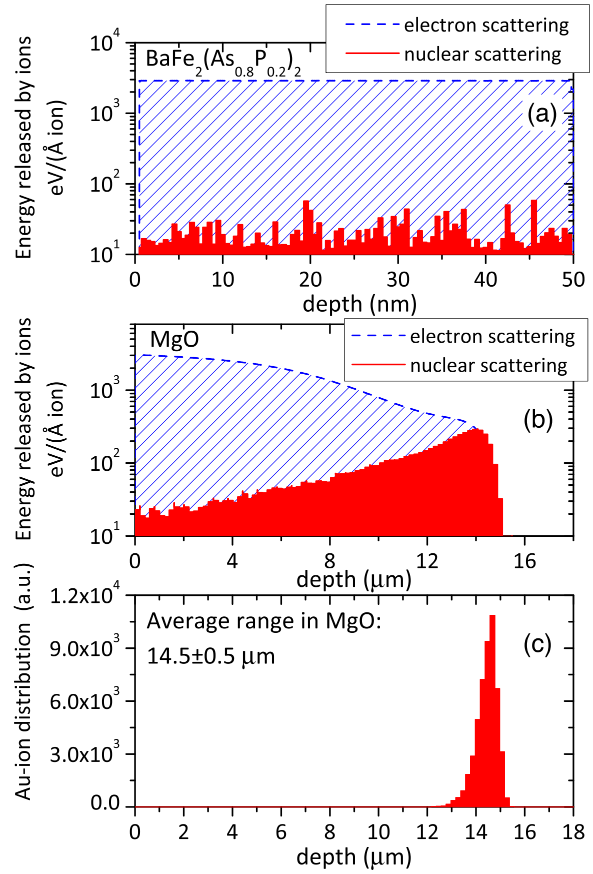

Fig. 1. Results of the simulations of energy release in the P-doped Ba-122 films and in the MgO substrate carried out with the SRIM code. (a) Energy loss (in eV/Å ion) in the superconducting film (50 nm thick) as a function of depth. The contribution of electron scattering (dashed line) and nuclear scattering (solid line) are shown. Due to the very small thickness of the film, the energy release is perfectly homogeneous, as is the distribution of defects. (b) Same as in (a), but for the MgO substrate. (c) Spatial distribution of the implanted ions: in average, they stop at a depth of 14.5 ± 0.5 μm in the substrate.

Because of the large energy loss and the ion implantation in the substrate, we expect the substrate damage to play a role which is not easy to single out. In this sense, some cautions must be used in comparing the results with those obtained in single crystals. In particular, because of the tight connection between the critical temperature and the tensile strain, the effect of irradiation on the substrate is expected to reverberate on the properties of the superconducting film, even if the latter is less directly affected by the energy release of the Au ions.

4. Experimental results

4.1. Effect of irradiation on the structure

As shown in Fig. 2a, the XRD spectra of the pristine films only display the 00ℓ peaks of $BaFe_2(As,P)_2$ besides the peaks from the substrate. As discussed in detail elsewhere [16] this indicates that these films are free from impurity phases, have a good crystalline quality and grow with the c axis perpendicular to the film surface. Φ scans of the 103 peak revealed an almost perfect fourfold symmetry