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Interaction of superconductivity and itinerant-electron magnetism: Critical fields of Ni/V superlattices

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We present experimental results showing an anomalous behavior in the critical fields of superconductor/magnet superlattices. Close to the superconducting transition temperature the critical-field anisotropy is reversed (i.e., the parallel critical field is smaller than the perpendicular critical field). As the temperature is reduced the anisotropy reverts to the normal behavior. This effect is attributed to the competing interaction between the itinerant magnetism in the magnetic (Ni) layer and the superconducting (V) layers.

The study of competing mechanisms, in particular the interaction of superconductivity and magnetism, is a problem of much current interest in condensed matter physics. The compound $ErRh_4B_4$ in which the local Er magnetic moments strongly interact with the superconductivity has been the subject of much research[1] especially regarding the interaction of ferromagnetism and superconductivity. The coexistence of antiferromagnetism and superconductivity has been studied extensively in the $SmRh_4B_4$ compound.[1,2] We present here the first experimental study relating to the interplay of superconductivity and itinerant ferromagnetism in vanadium/nickel superlattices.

Close to the superconducting transition temperature the critical-field anisotropy is reversed and as the temperature is lowered the anisotropy reverts to normal behavior.

Metallic superlattices allow independent control of the strength of the magnetic and superconducting couplings and therefore provide ideal model systems in which to test theoretical ideas. We have studied the interaction of the superconductivity in vanadium with the itinerant ferromagnet nickel. This system is unique in that (a) good quality, crystalline, and well segregated superlattices can be manufactured from Ni and V, and (b) the Curie temperature T_C of Ni can be decreased below the superconducting transition temperature T_s(~ 5.4 K) of V by decreasing the Ni thickness to the vicinity of 10 Å. In addition, a large observable effect on the superconducting properties results from keeping the thickness of the vanadium around its coherence length $\xi_v \sim 400$ Å.

The Ni/V multilayered samples were prepared using high-rate magnetron sputtering on room-temperature, single-crystal (0° and 90°) sapphire substrates.[3]

Extensive x-ray characterization of the samples was carried out on a two-circle diffractometer. The x-ray source was a Rigaku 2-kW Cu tube (Cu-$K\alpha$) with a graphite crystal analyzer. The crystallographic and chemical order[4] were obtained from θ-2θ and ω (rocking) scans in reflection and transmission geometries at small and large scattering angles. Figure 1 shows the x-ray diffraction results at high angles in the θ-2θ geometry for V(50 Å)/Ni(8 Å) superlattice. The separation of the peaks is easily related to the superlattice periodicity as described earlier,[3] and is in good quantitative agreement with results obtained from the preparation parameters. In addition, the small-angle x-ray data exhibit at least three modulation peaks showing conclusively the existence of modulation with a wavelength of 58 Å. The coherence length perpendicular to the layers determined from the full width at half maximum of θ-2θ scan at the first-order Bragg peak positions is ~ 1000 Å, and the undulations of the layers (*not* mosaic spreading) is ~ 1.5°. Detailed modeling and fitting to a large amount of additional x-ray data were performed based on both the kinematic and dynamical theories.[5,6]

The results show that the structure perpendicular to the substrate consists of well segregated fcc(111) Ni layers and bcc(110) V layers with a mosaic spread of ~ 5° and an order-disorder transition below 8 Å layer thickness. In the plane, the structure is polycrystalline with a grain size larger than 50 Å. Earlier,[7] ion mill Auger measurements have shown the oxygen or carbon contamination to be below detection limit beyond ~ 100 Å from the surface, in similarly prepared samples. Oxygen and carbon contamination is of serious concern since the V superconducting transition temperatures are drastically affected by slight contamination. To avoid this, long pump-down time, baking to ~ 100° C, and fast sputtering rates were required.

The magnetic properties of Ni films have been shown earlier to be strongly thickness dependent in similar fcc/bcc superlattices.[8] The magnetization and the Curie temperature

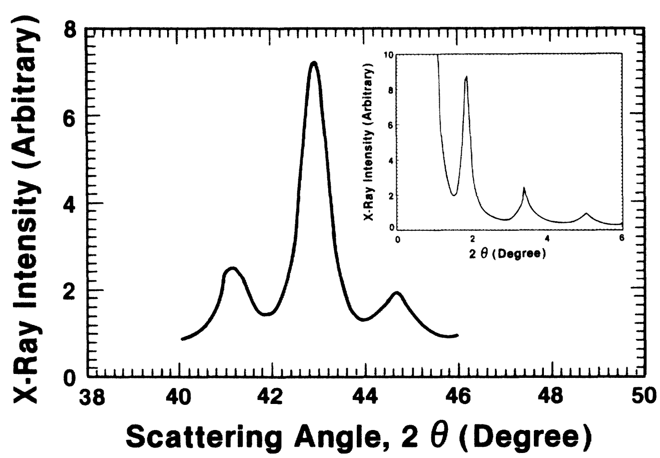

FIG. 1. High-angle θ-2θ x-ray diffraction of a V(50 Å)/Ni(8 Å). The inset shows small-angle θ-2θ diffraction results. Both graphs prove the existence of modulation structure at 58 Å as described in the text.

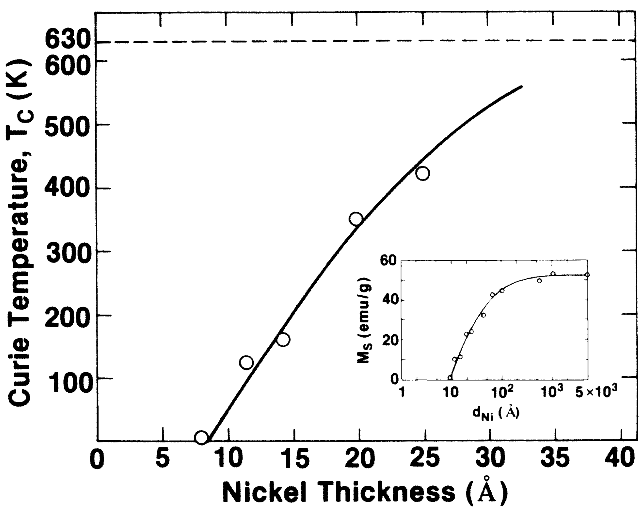

FIG. 2. The dependence of the Curie temperature vs nickel thickness, d_{Ni} in Mo/Ni superlattices, obtained from Arrot plots. Inset: dependence of the saturation magnetization on d_{Ni}. Lowest temperature for the measurements is 5 K.

(measured down to ~ 5 K) of Ni decrease with thickness to zero at about an average Ni thickness $d_{Ni} \simeq 8$ Å, as shown in Fig. 2 and its inset. Recent polarized neutron scattering[9] measurements have shown that Ni becomes nonferromagnetic in these superlattices below $d_{Ni} \sim 18$ Å, which is consistent with our result.

Two-dimensional superconductors ($d_V < \xi$) that are layered with insulators[10] or normal metals[11] exhibit the so-called "dimensional crossover" in their critical fields. As the layer separation decreases below the perpendicular coherence length, the critical-field temperature dependence changes from linear (three dimensional) to square-root-like (two dimensional).

The superconducting critical fields of magnetic-superconducting layered materials are very sensitive probes of the interaction between the two competing ordering phenomena as well as of dimensional behavior.[11] The critical fields were measured to a temperature of 1.1 K, up to fields of 5 T in the perpendicular and parallel directions and as a function of angle at fixed temperature for a large number (~ 40) of samples of thicknesses in the range 4 Å $\leq d_{Ni} \leq 100$ Å and 5 Å $\leq d_V \leq 10\,000$ Å. To avoid spurious results due to the nucleation of surface superconductivity the samples were always covered on both sides by Ni. The inset in Fig. 3 shows the temperature-dependent critical fields $H_{C2\parallel}(T)$ and $H_{C2\perp}(T)$ for a Ni(69 Å)/V(745 Å) superlattice. The behavior is typical of a three-dimensional (3D) superconductor with neither anisotropy nor surface superconductivity, i.e., $H_{C2\parallel}$ and $H_{C2\perp}$ show a linear temperature dependence. A thick (~ 1.15 μm) pure V sample exhibits identical behavior, i.e., the critical field is isotropic and linear as a function of temperature. In addition, the transition temperature ($T_C = 4.8$ K) is close to that of bulk V. If the V thickness is decreased close to its coherence length the T dependence becomes typical of two-dimensional (2D) superconductors as shown in Fig. 3 for the Ni(19 Å)/V(518 Å) sample. For this case, $H_{C2\parallel}$ shows a square-root-like temperature dependence, close to T_C whereas $H_{C2\perp}$ is still linear. These results are well understood in terms of the dimensional behavior of anisotropic superconductors.[11] The Ni in this region is sufficiently thick to decouple the V layers and therefore the behavior is simply controlled by the individual superconducting layers; for

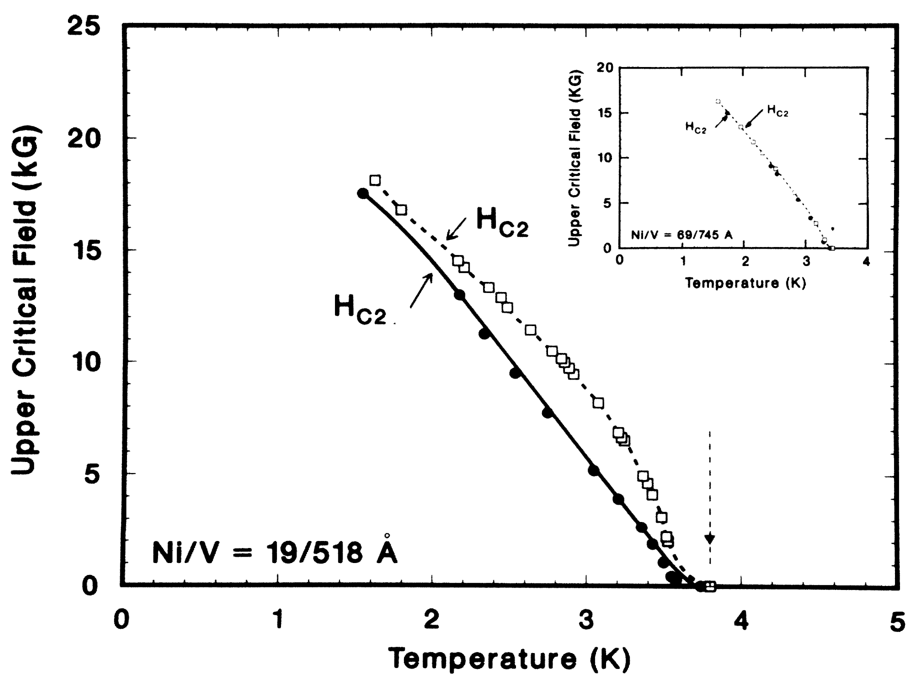

FIG. 3. Critical fields (measured from the midpoint of the transition) in the (□) parallel and (●) perpendicular direction for a two-dimensional sample. The arrow indicates T_s. Inset: critical fields in the (□) parallel and (●) perpendicular direction for a three-dimensional sample. The arrow indicates T_s. Typical transition widths (10%–90%) are less than the size of the dots.

large d_V the behavior is 3D, for small d_V the behavior is 2D. Note that $H_{C2\parallel} > H_{C2\perp}$ at all T.

The samples with the Ni thickness in the 10 Å range show quite interesting behavior. This is close to the thickness around which the magnetism in the itinerant Ni layers is weakened by the proximity of, or admixture with, the V layer. The anisotropy is reversed at high temperatures and then it reverts to the familiar $H_{C2\parallel} > H_{C2\perp}$ at low temperatures, as shown in Fig. 4 for one of the *typical* examples, a Ni(8 Å)/V(306 Å) sample. The temperature at which the anisotropy reverts is strongly dependent on the Ni thickness, for Ni thicknesses close to 10 Å. The extreme sensitivity of the critical-field anisotropy to the thickness of the Ni layer indicates that the effects reported here are mainly due to the weak magnetism present in the Ni (see Fig. 2) and in no way attributable to some subtle structural effects.

At this point, we would like to stress that the crossover in the anisotropy is extremely anomalous behavior and to our knowledge *never observed before* in any system. It should also be emphasized that the behavior described above is reproducible, systematic, and has been observed in a large number of samples. The details of the critical-field measurements, angular dependences, and transition temperatures for the rest of the samples will be the subject of a later publication.

The reason for these anomalies is not fully understood at the present time. Perhaps a complicated structure consisting of a complex multiconnected superconducting matrix in which paramagnetic or ferromagnetic Ni platelets are embedded could explain the results. We have not been able to reconcile our structural data with any of these type of models. Moreover, the extreme sensitivity of the superconductivity to changes in the Ni thickness gives a strong indication that the results are related to the magnetism in the Ni and not some subtle structural effect. The existence of anisotropy in the critical fields clearly shows that if there is an unknown complicated structure this has to be anisotropic. In addition, we do not know of any theoretical model of a complex system which could possibly explain the anomalous critical-field results presented here.

A theoretical idea has been advanced which relies on the interaction between superconductivity and the itinerant magnetism in the Ni.[12] As is well known, shape anisotropy in magnetic films tends to keep the magnetic moment in the direction parallel to the plane of the film. The magnetism is relatively weak in the temperature range close to T_s (assuming $T_C \sim T_s$) and therefore the whole sample is superconducting due to the proximity effect. However, the internal magnetic field has a larger parallel than perpendicular component due to the magnetic shape anisotropy. Therefore, $H_{C2\parallel} < H_{C2\perp}$ close to T_s. This effect is similar to that observed due to the electromagnetic effect in $ErRh_4B_4$ films.[13] At low temperature, the magnetism becomes strong enough to decouple the layers and therefore a dimensional crossover occurs to a 2D state with $H_{C2\parallel} > H_{C2\perp}$. Although this theoretical model is in qualitative accord with the experimental data, detailed calculations and fits are needed to ascertain whether it is operative here. Further polarized neutron scattering to study the magnetic moment distribution in the superconducting state, the influence of the structure and sharpness of interfaces, the interaction in other systems such as rare-earth/superconductor systems or antiferromagnet/superconductor systems would be of great interest.

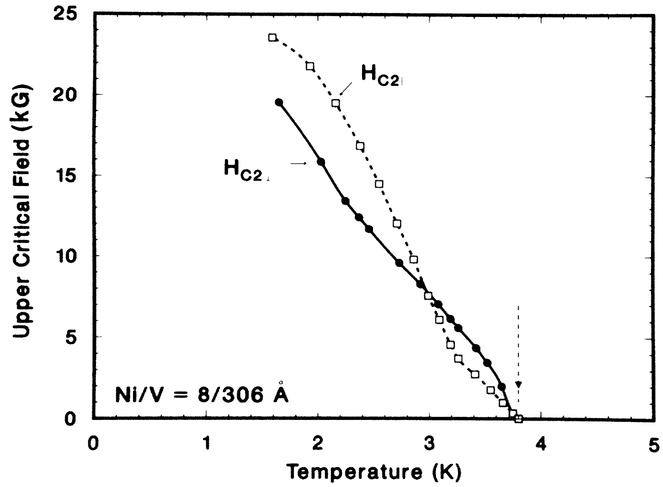

FIG. 4. Critical field (measured from the midpoint of the transition) in the (□) parallel and (●) perpendicular direction for one of the *typical* samples showing anomalous behavior. Notice that close to T_s the anisotropy is reversed and a crossover occurs to the normal behavior at low temperature. Typical transition widths are less than the size of the dots.

In summary, we have studied the critical-field anisotropies for the superlattice of superconducting (V)/magnetic (Ni) metals for thicknesses in which the superconducting transition temperature is close to the magnetic transition temperature. The critical-field anisotropy is anomalous in that close to the superconducting transition temperature and the parallel critical field is smaller than the perpendicular critical field while at low temperature it reverts to the ordinary anisotropy. This behavior appears to be due to the interaction of the magnetism in the Ni films with the superconductivity in the V layers. We hope that this work will stimulate further work in the theory of these systems and in the use of superlattices as model systems to study competing mechanisms.

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