PHYSICAL REVIEW B VOLUME 41, NUMBER 18 15 JUNE 1990-II

Brief Reports

Brief Reports are short papers which report on completed research which, while meeting the usual **Physical Review** *standards of scientific quality, does not warrant a regular article. (Addenda to papers previously published in the* **Physical Review** *by the same authors are included in Brief Reports.) A Brief Report may be no longer than* $3\frac{1}{2}$ *printed pages and must be accompanied by an abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

NMR study of interface structure in epitaxial Co-Cu superlattices

K. Le Dang and P. Veillet
Institut d'Électronique Fondamentale, Bâtiment 220, Université de Paris–XI, 91405 Orsay CEDEX, France

Hui He, F. J. Lamelas, C. H. Lee, and Roy Clarke
Department of Physics, Randall Laboratory, The University of Michigan, Ann Arbor, Michigan 48109-1120

(Received 8 February 1990; revised manuscript received 2 April 1990)

Co spin-echo NMR has been measured in a series of Co-Cu superlattices grown by molecular-beam epitaxy. The results demonstrate the usefulness of NMR as a probe of interface structure in cobalt-based superlattices. The hyperfine satellites uniquely identify interfacial Co atoms with three Cu nearest neighbors, indicating the presence of atomically abrupt interfaces. This is the first NMR observation of Co atoms at interfaces in a superlattice sample. The measurements also confirm that the Co layers have fcc stacking symmetry with an epitaxially induced in-plane lattice expansion.

Cobalt and copper are interesting elements for superlattice studies because their fcc lattice parameters have nearly the same value, i.e., 3.55 and 3.61 Å, respectively. In a previous study[1] we have described the molecular-beam-epitaxy (MBE) growth of epitaxially oriented fcc Co-Cu superlattices. These samples, along with Co-Au superlattices, have also been the subject of a recent study of magnetic anisotropies.[2] We note that magnetic properties such as perpendicular anisotropy are sensitive to interface structure, yet the interfaces in Co superlattices are difficult to characterize via conventional transmission-electron-microscopy (TEM) and x-ray techniques. This is particularly true in the case of Co and Cu, due to the poor optical contrast which arises from their nearly adjacent positions in the Periodic Table. In this Brief Report we will show that the spin-echo NMR technique is very useful as a probe of interface structure, given the sensitivity of the NMR spectrum to the local environment of the Co atom. The spectrum is also quite sensitive to epitaxial strains in the Co layers, since a change in the lattice parameter will have a marked effect on the Co hyperfine field.

The Co-Cu superlattices were prepared in a Vacuum Generators VG-80 MBE system on annealed GaAs(110) substrates.[1] All of the samples contained Cu layers of thickness 23±3 Å, with a total superlattice thickness of 1500 Å. The Co-layer thicknesses ranged from 20 Å (~10 monolayers) to 65 Å (32 monolayers). Reflected high-energy electron-diffraction and x-ray-scattering measurements[1] show that the superlattices consist of layers of close-packed planes which are epitaxially oriented within the growth plane. Detailed x-ray studies have also shown that the Co structure is essentially fcc for layer thicknesses of up to 65 Å.[1,3]

Traditionally, the superlattice composition profile normal to the layers is probed by out-of-plane x-ray scans where the scattering vector is perpendicular to the layers. In this approach the sharpness of the interfaces is determined by an analysis of the superlattice satellite intensities.[4] However, the Co-Cu system is difficult to analyze via x-ray techniques because in this case the superlattice satellites arise from a lattice *spacing* modulation rather than a composition modulation. This effect is shown in Fig. 1, where we have plotted the out-of-plane (lll) x-ray-scattering intensities calculated using a step model with ideally abrupt interfaces. We find that a superlattice satellite, approximately 2 orders of magnitude weaker than the main (111) peak, occurs when the Co and Cu layers have bulk close-packed plane spacings of 2.035 and 2.087 Å, respectively. However, if both layers are given the lattice parameter of Co, the superlattice satellite vanishes. In the Co-Cu system the x-ray data are insensitive to *composition* modulation, and thus the NMR data are invaluable for characterizing the interface.

The NMR measurements were carried out at liquid-helium temperature using a variable-frequency spin-echo apparatus.[5] The superlattice samples were mounted in a Dewar tail around which was fitted an exciting coil, such that the rf field was parallel to the film plane. The surface area of the film ranged from 0.3 to 0.8 cm^2.

The resonance frequency of the main NMR peak is 215 MHz (Fig. 2), as compared to 217.5 MHz in bulk fcc cobalt. This decrease in frequency is probably due to an expansion of the cobalt lattice in Co-Cu superlattices. Indeed, the resonance frequency has been found to increase with pressure by 128 kHz/kbar in bulk fcc cobalt.[6]

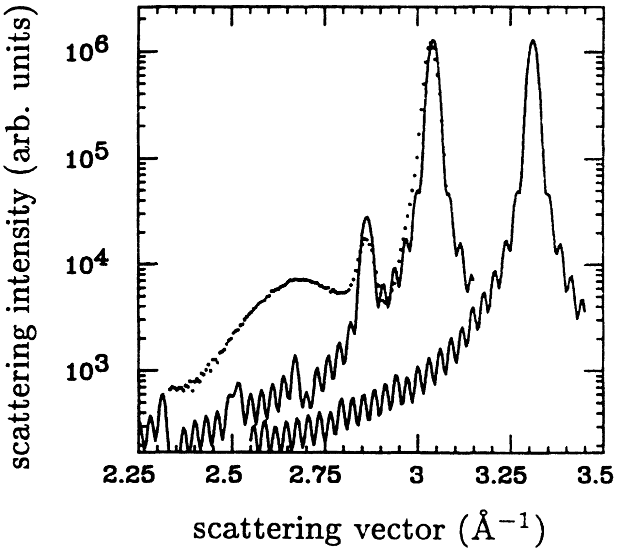

FIG. 1. Measured out-of-plane (*lll*) x-ray-scattering intensities (points), plotted with a step-model calculation (solid curve) of a six-bilayer superlattice with Co and Cu layers having bulk lattice parameters. The second calculation (displaced to the right for clarity) was performed with Co and Cu layers each with the bulk lattice parameter of Cu. Both calculations included seven and ten atomic layers, respectively, of Co and Cu with automatically abrupt interfaces. The broad peak near 2.7 Å^{-1} (in the measured data) arises from the (111) peak of a 20-Å protective Au cap on the superlattices.

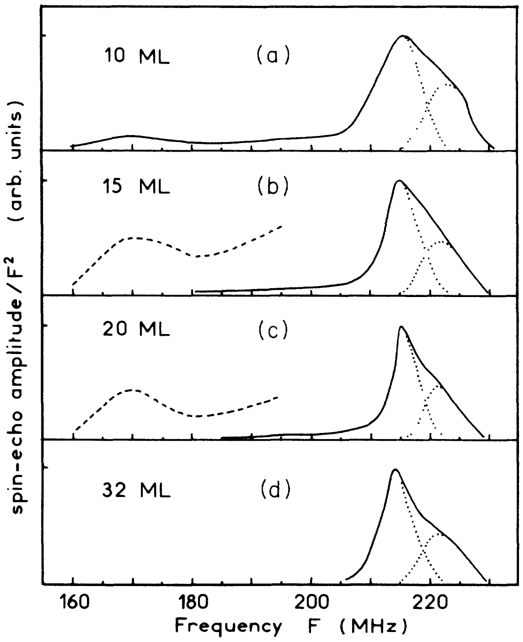

FIG. 2. Co spin-echo spectra at $T=2$ K in Co-Cu superlattices with Co-layer thicknesses (in monolayers) of (a) 10, (b) 15, (c) 20, and (d) 32. The main peak was separated into two parts (dotted lines), showing the effect of stacking faults. The dashed curves correspond to a tenfold magnification of the vertical scale.

Assuming that the relative change in lattice parameter, $\Delta a/a$, remains small, the pressure dependence yields a NMR freqency shift of $\delta\nu(\mathrm{MHz})=-776(\Delta a/a)$. This value indicates that our observed frequency shift of -2.5 MHz corresponds to a Co lattice expansion of 0.3%. In comparison, our x-ray measurements[1] yield a Co lattice expansion of $\gtrsim 1\%$. The difference between these two strain values might be interpreted as follows. The NMR peak at 215 MHz arises from Co atoms where all 12 of the nearest neighbors are Co, i.e., from Co atoms within the interior of the Co layers. The x-ray measurement, on the other hand, averages over the entire superlattice, including the Cu layers and those Co atoms at the interfaces. The lower strain value indicated by the NMR measurements implies that a strain gradient exists within the superlattice, with the centers of the Co layers partially relaxed towards the bulk Co lattice spacing.

The high-frequency wing of the main peak can be ascribed, like the 223-MHz satellite line in bulk fcc Co, to stacking faults.[7] This effect can be understood by considering a close-packed sequence such as *ABCBCABC* containing a single stacking fault, where the Co atoms in *two* planes are no longer in a cubic environment. The fraction of planes in a hcp environment, proportional to the integrated signal intensity around the 223-MHz shoulder (Fig. 2), was estimated to be $\sim 20\%$, in agreement with x-ray-scattering results.[1]

The most interesting feature of the resonance spectrum is the low-frequency tail, which contains a small peak near 170 MHz and vanishes below 160 MHz. For the sample with the thickest (32 monolayer) Co layers, which has the smallest area (0.3 cm^2), this signal is masked by noise [Fig. 2(d)]. For the other samples the low-frequency signal intensity increases as the Co-layer thickness decreases, revealing that it is interfacial in origin [Figs. 2(a)–2(c)].

In order to use this feature as a quantitative probe of Co atoms at interfaces, the NMR Co satellite lines were measured for a dilute Co-Cu alloy (3% Cu). The resonance peaks P_1, P_2, and P_3 appearing at 199, 180, and 163 MHz are associated with Co atoms having one, two, and three Cu nearest neighbors, respectively (Fig. 3). These peaks occur at nearly regular intervals from the main peak at 217.4 MHz. This result shows that the influence of the nearest neighbors on the Co hyperfine field is predominant. The relative intensities of the NMR spectrum of the alloy (Fig. 3) are consistent with a ran-

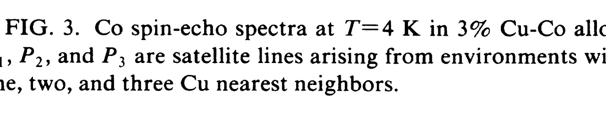

FIG. 3. Co spin-echo spectra at $T=4$ K in 3% Cu-Co alloy. P_1, P_2, and P_3 are satellite lines arising from environments with one, two, and three Cu nearest neighbors.

dom distribution of Cu impurities. The statistical weight of Co environments with n Cu atoms among 12 nearest neighbors is given by the expression

$$P_n = \binom{n}{12} c^n (1-c)^{12-n} , \qquad (1)$$

where $\binom{n}{12}$ is the binomial coefficient and c is the impurity concentration. The relative signal intensities in the 3% alloy sample (Fig. 3) and in another alloy with $c = 2\%$ are in agreement with a random distribution.

In superlattices containing close-packed (111) planes, each Co atom at the interface is neighbored by three Cu atoms. These atoms, unlike those in the random P_3 environment in bulk material, are in an axial crystal field. Their hyperfine field is expected to be isotropic within the film plane where the magnetization is lying. Owing to the preponderant effect of the nearest neighbors on the Co hyperfine field, the line at 170 MHz (Fig. 2), a frequency slightly higher than ν_{P_3} (163 MHz Fig. 3), can be ascribed to the Co atoms at the (111) interfaces. In addition, the superlattice spectra contain very weak intensities at ν_{P_1} and ν_{P_2}. The measured intensities are similar to those expected in the ideal case of a superlattice with atomically abrupt interfaces, which would contain Co atoms in only two environments: either within the Co layers (with 12 Co nearest neighbors) or at the interfaces (with nine Co neighbors and three Cu neighbors). In this ideal case, peaks would occur only near 220 MHz and near ν_{P_3}. On the other hand, rough or alloyed interfaces would contain many Co atoms with one or two Cu near neighbors, leading to significant intensities at ν_{P_1} and ν_{P_2}. From the signal intensity at 200 MHz (ν_{P_1}) in the superlattice spectra, we estimate that copper diffusion in cobalt layers is less than 0.4%, 0.6%, and 0.9% for the samples with Co-layer thicknesses of 20, 15, and 10 monolayers. These concentrations vary roughly as the inverse thickness, i.e., as the total interface area, as expected. Finally, we note that the integrated intensity in the range 160–200 MHz (near ν_{P_3}) arises primarily from Co atoms at the interfaces. Its ratio, with respect to the principal peak at 215 MHz, is in all cases [Figs. 2(a)–2(c)] consistent with the ratio of two interfacial Co monolayers to $n-2$ inner monolayers, where n is the total number of Co monolayers in each layer.

In conclusion, we have shown that the spin-echo NMR technique can be used to unambiguously identify the presence of atomically abrupt interfaces in Co-Cu superlattices. This technique may be especially useful in cases where an analysis of the interfaces by traditional techniques proves difficult. In addition, the NMR technique can differentiate between hcp and fcc stacking sequences within the Co layers, and can be used to estimate epitaxial strains within the layers. We expect that NMR will be useful as a probe in Co superlattice systems where the properties of the interface are critical.

The Institut d'Électronique Fondamentale is a Unite de Recherche associée au Centre National de la Recherche Scientifique, No. 22. Work at the University of Michigan was supported in part by the U.S. National Science Foundation under Grant No. DMR-88-05156.

---

[1] F. J. Lamelas, C. H. Lee, Hui He, W. Vavra, and Roy Clarke, Phys. Rev. B **40**, 5837 (1989).

[2] C. H. Lee, Hui He, F. J. Lamelas, W. Vavra, C. Uher, and Roy Clarke, Phys. Rev. B (to be published).

[3] F. J. Lamelas, Ph.D. thesis, The University of Michigan, Ann Arbor, 1990.

[4] See, for example, D. B. McWhan, in *Physics, Fabrication, and Applications of Multilayered Structures*, edited by P. Dhez and C. Weisbuch (Plenum, New York, 1988).

[5] A related NMR study of a Au-Co-Au sandwich, concerned with structure *within* the Co layer, was reported by K. Le Dang, P. Veillet, C. Chappert, P. Beauvillain, and D. Renard, J. Phys. F **16**, L109 (1986).

[6] R. V. Jones and I. P. Kaminov, Bull. Am. Phys. Soc. **5**, 175 (1960).

[7] R. Street, D. S. Rodbell, and W. L. Roth, Phys. Rev. **121**, 84 (1961).