Direct Evidence of Spin Polarization Oscillations in the Cu Layers of Fe/Cu Multilayers Observed by NMR

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Evidence of the existence and distribution of an induced spin polarization of the conduction electrons in the Cu layers of Fe/Cu multilayers has been obtained by nuclear magnetic resonance (NMR). Fine structure associated with the spin-echo signal of ^{63}Cu and ^{65}Cu nuclei shows that the spin polarization and an associated exchange field oscillate in sign, similar to the characteristics of the RKKY interaction. However, an accurate determination of the period of the oscillations cannot be determined solely from the NMR data at the present time.

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In recent years, the coupling of the ferromagnetic layers in multilayer films through the nonmagnetic metallic layers has received a great deal of attention [1,2]. Magnetic layers separated by a nonmagnetic layer can be aligned ferromagnetically (FM) or antiferromagnetically (AFM) with respect to each other. In a number of systems, such as Fe/Cr, Co/Cu, Fe/Cu, etc. [3–6], the interlayer coupling oscillates between FM and AFM as a function of the thickness of the nonmagnetic layer. This oscillatory behavior has been demonstrated in many systems by measurements of the magnetism, the magnetoresistance, etc. A reasonable assumption then is that the spin polarization of the conduction electrons in the nonmagnetic layers displays some type of spatially oscillatory behavior. In fact, evidence of such an oscillation in the nonmagnetic overlayer on a ferromagnetic substrate has been obtained by spin-polarized electron-energy-loss spectroscopy [7] and by scanning electron microscopy with polarization analysis [8]. Similar observations have also been made in sandwiches from the spin-resolved tunneling current [9]. Furthermore, indirect evidence of spin polarization from magnetic and magneto-optical measurements in multilayers has been obtained [10,11]. However, direct evidence of spin polarization oscillations in the nonmagnetic layers has not been previously reported in multilayer films.

Nuclear magnetic resonance (NMR) provides a direct way to investigate the spin polarization of conduction electrons by means of the Knight shift. Lang *et al.* [12] and Boyce and Slichter [13] used a spin-echo technique on Cu nuclei to study the electron-spin density in CuCo and CuFe dilute alloys. Both of them found several resonant "satellite" peaks corresponding to Cu atoms which were the first, second, and farther nearest neighbors of the Co or Fe atoms, and the extra Knight shift $\Delta K/K$, due to the spin polarization of the conduction electrons, showed oscillations between positive and negative values.

Our previous paper [14] reported preliminary results of our observation of an additional Knight-shift oscillation in the NMR or ^{63}Cu nuclei in Fe/Cu multilayers. We report here further NMR results on Fe/Cu multilayers from both ^{63}Cu and ^{65}Cu nuclei, confirming the spin polarization oscillation of the conduction electrons in the Cu layers.

The Fe/Cu multilayers were prepared by rf sputtering in a system with a base pressure of 3×10^{-6} Torr. An argon gas pressure of 7 mTorr was used during deposition. The compositionally modulated structure was achieved by rotating the water-cooled substrate and alternately expos-

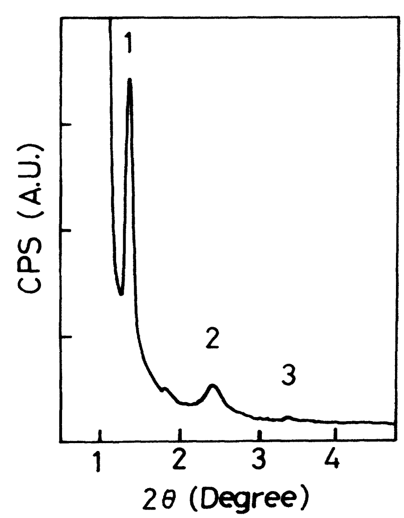

FIG. 1. Low-angle x-ray diffraction pattern of a Fe/Cu multilayer on a glass substrate.

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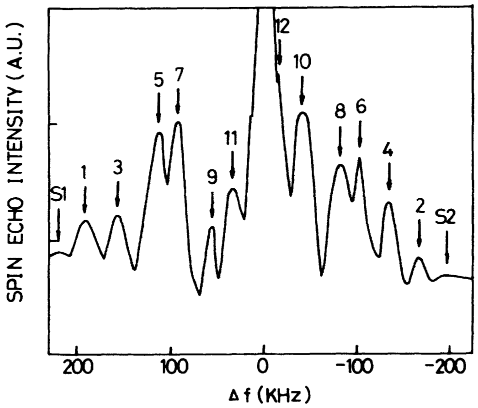

FIG. 2. Spin-echo spectrum of ^{63}Cu nuclei in a [Fe(60 Å)/Cu(25 Å)]$_{50}$ multilayer deposited on a glass substrate.

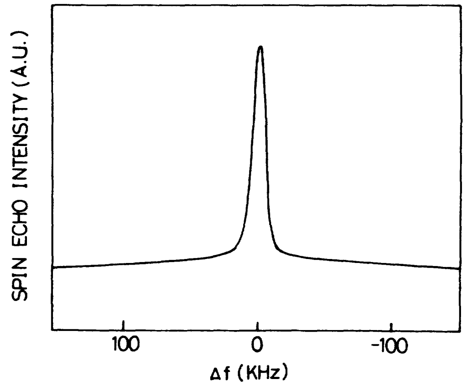

FIG. 4. Spin-echo spectrum of ^{63}Cu nuclei in a single copper film with a thickness of 400 nm.

ing it to each of the two targets. The deposition rates of Fe and Cu were 2.0 and 1.1 Å/s, respectively. The Fe thickness was 60 Å, the Cu thickness was 25 Å, and the number of periods was 50. Both glass and Kapton substrates were used. From low- and high-angle x-ray diffraction, the films were confirmed to be well periodically layered, and polycrystalline with a (111) texture for the Cu. Figure 1 shows the low-angle x-ray diffraction pattern of a Fe/Cu multilayer film deposited on a glass substrate.

The NMR measurements were performed on a Bruker MSL-300 NMR spectrometer at room temperature. High sensitivity was achieved by using a pulse-echo and Fourier-transformation method. Signal averaging was carried out using typically 10^6 to 10^7 individual traces. A constant magnetic field of 7 T was applied parallel to the film plane.

For the empty probe, a relatively strong NMR signal from both ^{63}Cu and ^{65}Cu nuclei was observed due to some material in the probe containing copper. The ^{63}Cu signal occurred at a frequency of 79.74320 MHz, the same as that of a pure copper film or copper powder used as a reference. Both gave the standard Knight shift of 0.233% when compared with a $CuCl_2$ powder sample. Figures 2 and 3, respectively, show the NMR spectra of ^{63}Cu nuclei in [Fe(60 Å)/Cu(25 Å)]$_{50}$ multilayers deposited on substrates of glass and Kapton, for which the resonant frequency of the pure copper film is taken as the zero point. It can be seen that the spectra of the two samples on glass and Kapton are quite similar. A group of satellite peaks is distributed on both sides of the main resonant peak, which is due mainly to the signal coming from the probe.

In order to confirm that the multiple-peak signals reflect intrinsic NMR transitions within the multilayer samples, instead of a spurious signal, we conducted two tests. First, we measured the NMR spectrum of ^{63}Cu in a single copper film of 400 nm thickness. As shown in Fig. 4, this produced a clean signal peak with the Knight shift mentioned above. Second, we performed an NMR measurement on the ^{65}Cu nuclei (shown in Fig. 5) in the same sample used for Fig. 2. This isotope also showed a multiple-peak spectrum, in which the number and posi-

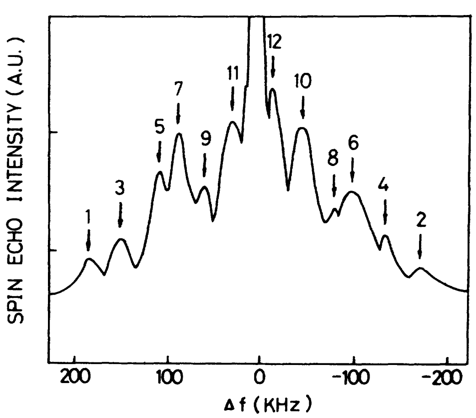

FIG. 3. Spin-echo spectrum of ^{63}Cu nuclei in a [Fe(60 Å)/Cu(25 Å)]$_{50}$ multilayer deposited on a substrate of Kapton.

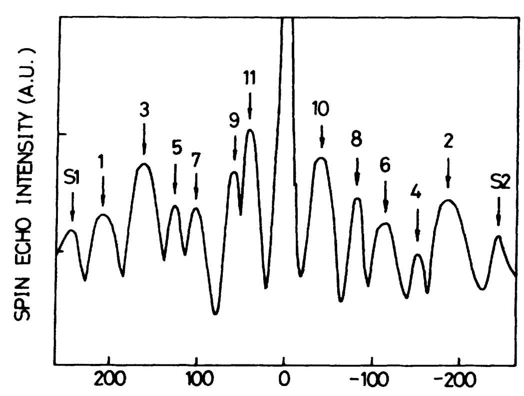

FIG. 5. Spin-echo spectrum of ^{65}Cu nuclei in the same sample used for Fig. 2.

TABLE I. The exchange fields (in kOe) corresponding to the peaks in Fig. 2.

H_{s1}	H_{s2}	H_1	H_2	H_3	H_4	H_5	H_6	H_7	H_8	H_9	H_{10}	H_{11}	H_{12}
⋯	⋯	72	−62	58	−50	43	−38	36	−30	21	−15	14	−5

tions of the satellite peaks are similar to those of the ^{63}Cu spectrum, although the intensities of the peaks are different.

A multiple-peak NMR spectrum could be caused by perturbations of the quadrupolar interaction due to a distorted cubic lattice within the Cu layers. However, since the spin quantum numbers of both ^{63}Cu and ^{65}Cu nuclei are $\frac{3}{2}$, the quadrupolar interaction should give rise at most to only two symmetrical satellite peaks, or a mere broadening of the single peak if the strain varies continuously. Thus, we conclude that the origin of the satellite peaks with a multiplicity up to 6 on both sides of the main peak is more likely a result of a nonuniform spin polarization of the conduction electrons in the Cu layers caused by exchange coupling to the Fe. From the positions of the multiple peaks, we can obtain the effective magnetic field experienced by the nuclei, which we have called the exchange field H_{ex}.

It is well known that the NMR resonance frequency in metals can be expressed as

$$f_0=\gamma H_0(1+K)=\gamma H_0(1+\Delta H/H_0)\,, \tag{1}$$

where γ is the gyromagnetic ratio of the nuclei under study ($\gamma=11.285$ MHz/T for ^{63}Cu and 12.089 MHz/T for ^{65}Cu), K is the Knight shift of the metal, and ΔH is the Fermi contact field experienced by the nuclei due to the spin polarization of the conduction electrons caused by the external magnetic field H_0. In our multilayer films, the additional spin polarization in the Cu layers, caused by the exchange field H_{ex}, leads to a modified expression for the resonant frequency containing a new term representing the additional Knight shift:

$$\begin{aligned} f&=f_0+\Delta f=\gamma H_0(1+K+\Delta K)\\ &=\gamma H_0[1+\Delta H/H_0+(\Delta H/H_0)H_{ex}/H_0]\,. \end{aligned} \tag{2}$$

Thus, the exchange field in the Cu layers associated with any particular peak is given by

$$H_{ex}=\Delta f/\gamma K\,, \tag{3}$$

where Δf is the additional Knight-shift frequency, which can be measured directly from the NMR spectra in Figs. 2, 3, or 5.

Tables I–III give the calculated values of H_{ex} from Eq. (3) above. The maximum value of H_{ex} is about 82 kOe in the ^{65}Cu spectrum. This may correspond to the Cu atoms nearest to the Fe interfaces. It is interesting that with the decrease of the absolute value of the exchange field, positive and negative values appear alternately as shown in the tables. This is exactly what one expects from an induced spin polarization which extends into the Cu as a spatially damped oscillation from the Fe-Cu interface. A comparison of the corresponding values of the exchange field H_{ex} in Tables I and II, for the two samples with different substrates but a similar layered structure, shows good agreement with a maximum difference of about 10% or less, while the agreement between the values of H_{ex} in Tables I and III, for the same sample but with the different isotopes of ^{63}Cu and ^{65}Cu, is better. This difference is only about 5% or less. With the limited range of sample thicknesses we have studied to date, it is not possible from the NMR data alone to determine the spatial locations within the samples that give rise to the NMR peaks and, thereby, the oscillation period of the electron polarization and H_{ex}. If we make the simplest assumption that the regions producing the NMR peaks are equally spaced along the normal to the Cu layer plane, the total number of oscillations is about 6, giving an average period of 4-5 Å within the Cu layer. This is a shorter period than we would expect.

As is known from RKKY theory, the strength of the magnetic coupling between individual atoms a distance R apart varies as $R^{-3}\cos(2k_FR)$. In layered systems, the interaction between magnetic layers decreases asymptotically as the inverse square of the spacer thickness [15,16]. The periodicity of the spin-density oscillation is thus determined by k_F, the wave vector of the conduction electrons at the Fermi level. This is in general about 2 lattice spacings or monolayers (ML). An oscillation with a short period of 2 ML has been predicted by a theoretical calculation of the coupling [17]. Also, recent experiments on the layer by layer spin polarization in epitaxial Cr overlayers on Fe(100) [7,8] have been conducted, in which the oscillation period is about 2 atomic layers.

However, our estimated short period of 4-5 Å in Fe/Cu multilayers with a (111) texture for the Cu does not coincide with either the theoretical prediction (~9 Å) along the same Cu(111) direction [18] or with other experimental data (~12 Å) on sputtered Fe/Cu, mainly (111) oriented [6]. Bruno and Chappert [18] have presented a model, based on the RKKY interaction, to explain the oscillation period of the interlayer coupling

TABLE II. The exchange fields (in kOe) corresponding to the peaks in Fig. 3.

H_{s1}	H_{s2}	H_1	H_2	H_3	H_4	H_5	H_6	H_7	H_8	H_9	H_{10}	H_{11}	H_{12}
⋯	⋯	69	−64	57	−50	40	−38	32	−30	22	−17	13	−5

TABLE III. The exchange fields (in kOe) corresponding to the peaks in Fig. 5.

H_{s1}	H_{s2}	H_1	H_2	H_3	H_4	H_5	H_6	H_7	H_8	H_9	H_{10}	H_{11}	H_{12}
82	−78	70	−61	56	−51	43	−38	35	−29	20	−15	14	···

for fcc (111), (001), and (110) grown systems (Cu, Ag, Au), in which they consider the discreteness of the spacer thickness and the moment distribution within the ferromagnetic layers. Their calculations show both short (2.6 ML) and long (5.9 ML) periods for a Cu spacer in the (001) orientation, but only a long period (4.5 ML) for the (111) orientation. Short- and long-period oscillations are observed in Fe/Cr/Fe [19,20], Fe/Mo/Fe [21], Co/Cu/Co, Fe/Cu/Fe [22], Fe/Al/Fe, and Fe/Au/Fe [23] multilayered structures.

The reason for the discrepancy between our estimated period and the above theoretical and experimental results is not clear. It should be emphasized again that with our present NMR data, the regions producing the NMR peaks cannot be localized, and thus the period cannot be determined unambiguously. We have tried to fit our data for H_{ex} by a functional form whose amplitude decreases as $z^{-\gamma}$ (z is the distance from the magnetic interface and γ may be 1, $\frac{3}{2}$, or 2 [18,24]) by taking as variable parameters the amplitude and phase of the oscillation. We found the estimated period to be still ambiguous. Perhaps we are observing interference between signals coming from crystallites having different orientations. More work is required at this point. We have begun studying a series of samples having a range of different Cu thicknesses in an effort to better localize the regions within the Cu layers which contribute to the individual peaks of the NMR signal. It would also be very useful to investigate epitaxially deposited material and look for any orientational dependence of the Cu layers on the NMR signal characteristics and the inferred periodicity of the spin oscillations.

In summary, $[Fe(60\ Å)/Cu(25\ Å)]_{50}$ compositionally modulated films have been studied by NMR at room temperature. We have interpreted the appearance of additional satellite peaks accompanying the main NMR lines for the ^{63}Cu and ^{65}Cu nuclei as due to an oscillatory Knight shift resulting from a spatially dependent oscillation of the spin polarization of the conduction electrons in the Cu layers. If this interpretation is correct, we have direct evidence for the first time in a multilayer sample of an oscillatory exchange field and spin polarization within the conduction electrons of the nonmagnetic component, which can play an important role in the interlayer coupling between the ferromagnetic layers of the film.

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