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Interlayer exchange coupling in fine-layered Fe/Au superlattices

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Interlayer exchange coupling (IEC) in Fe (n ML)/Au (n ML) superlattices (SL's) (ML's= monolayers, $n=1$ to 5) has been examined by Brillouin scattering from spin waves at 300 K. The IEC was found to be ferromagnetic for all n, but the IEC strength J_{i} exhibits oscillatory behavior: it is large for even ML's and small for odd ML's. For $n=1$ which correponds to an ordered alloy with the $L1_0$ structure, we obtain $J_{\mathrm{i}}=43.6\pm5.0$ mJ/m^2. *Ab initio* calculation of J_{i} by the self-consistent full-potential linearized augumented-plane-wave method gives a good agreement with the experimental values, except for $n=3$ where an antiferromagnetic ground state is predicted.

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The conventional system for the investigation of interlayer exchange coupling (IEC) is ''trilayers'' which consist of two ferromagnetic films with thicknesses on the order of a few 10 nm separated by interlayers with thicknesses more than a few monolayers (ML). Here, it is mostly assumed that the asymptotic limit is valid, i.e., the coupling is independent of the ferromagnetic layer thickness and the oscillation periods as a function of the interlayer thickness are given by certain distances, called calipers, in the Fermi surface of the interlayer material. In the case of Au interlayers with surface normal parallel to a [100] direction there are two such calipers, given by the extremal distances across the ''dogs bone orbit'' (DBO) in the Fermi surface of Au in the [100] direction. The experiments which have so far been performed for IEC in the Fe/Au/Fe trilayers grown on Ag buffered GaAs substrates[1] and on Fe whiskers[2] satisfy well the asymptotic limit and show clearly the expected oscillations with periods as given by the calipers of the DBO. For the samples on the whiskers the smallest Au interlayer thickness for which antiferromagnetic coupling could be detected was around 4 ML, with a surprisingly large coupling strength of ~1 mJ/m^2.[2] For the samples on the Ag buffer below 5 ML only ferromagnetic coupling could be identified, with increasing strength towards smaller Au thickness and some superimposed wiggles which indicated further oscillations of the coupling.[1]

Asymptotic limit means that the interlayer thickness should be large as compared to the Fermi wavelength, which in metals is of the order of the atomic distances. For thicknesses below this limit, IEC cannot be described by explicit formulas, but has to be treated numerically, for example *ab initio* by the self-consistent full-potential linearized augumented-plane-wave (FLAPW) method. This is one of the reasons why so far coupling across very thin interlayers has not been explored very much. Other reasons come from experimental difficulties. It is clear that for extremely thin interlayers ''true'' coupling across the interlayer material can easily be obscured by the presence of bridges of ferromagnetic material. Since direct exchange is by orders of magnitude larger than IEC, any direct contact of the ferromagnetic layers will tend to dominate the resultant coupling in the area of the sample where it occurs.

The MBE preparations of the Fe (n ML)/Au (n ML) ''fine layered'' superlattices (FLSL's) on MgO (001) substrates with $n=1$ to 5 were described elsewhere.[3–5] Total number of Fe and Au atomic planes were kept constant. Here, we use the term of FLSL for SL with layer thicknesses comparable or smaller than the Fermi wavelength. The Fe(1 ML)/Au(1 ML) FLSL corresponds to the ordered alloys with the $L1_0$ structure which exist in the the equilibrium phase diagram for Fe_1Pt_1 alloy but not in the Fe_1Au_1 alloy. In the Fe(1 ML)/Au(1 ML) FLSL it can only be obtained by artificial layering as in the present case. The structural and magnetic properties of the Fe/Au FLSL's were examined through x-ray diffraction (XRD), superconducting quantum interference device (SQUID) magnetometry, ferromagnetic resonance, and magneto-optical Kerr (MOK) effect. These efforts revealed a strongly enhanced magnetic moment of $2.8\mu_{\mathrm{B}}$ per Fe atom as compared to $2.2\mu_{\mathrm{B}}$ in the bulk,[4] oscillatory behavior of the lattice spacing, inplane and perpendicular magnetic anisotropies,[4,6] and MOK spectra which indicate changes in electronic structure.[7] Since the Fe-Au system is a typical model system, many *ab initio* studies have been performed on the magnetic and structural properties.[8–10]

Brillouin scattering (BS) has become a standard method for the investigation of magnetic thin-film structures,[11] in particular IEC in trilayers. In our preliminary BS work on Fe(2 ML)/Au(2 ML) SL with 50 periods,[12] we observed scattering from standing spin waves (SSW's) which allowed us to determine the IEC constant J_{i}. We have further continued our BS determination of the IEC in the Fe/Au FLSL's at room temperature. Formally, one can describe the IEC in the FLSL's in the same way as in the trilayers, but substantially there are appreciable differences. From the works on the

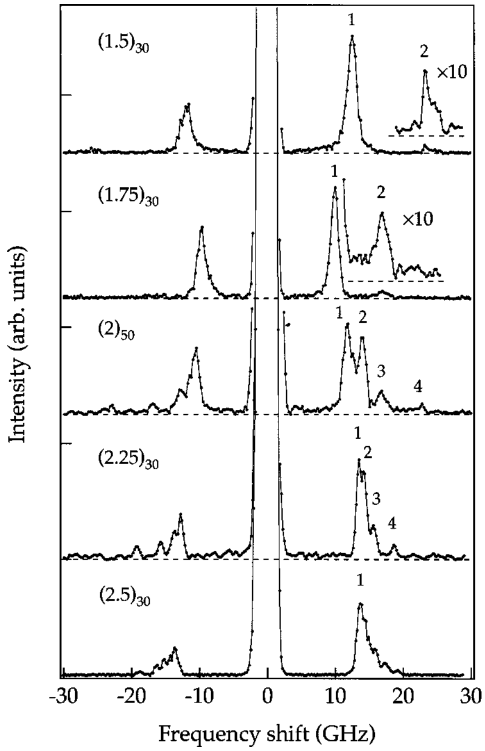

FIG. 1. BS spectra observed from ($2 \pm \delta$ ML) SL's ($\delta = 0.25$ and 0.5) at $H = 0.3$ T with $Q_S = 0.61 \times 10^7$ m^{-1}. Total thickness of each SL is 14.5, 17.6, 36.4, 25.6, and 26.9 nm from top to bottom. The labels 1 to 4 stand for the SSW mode number.

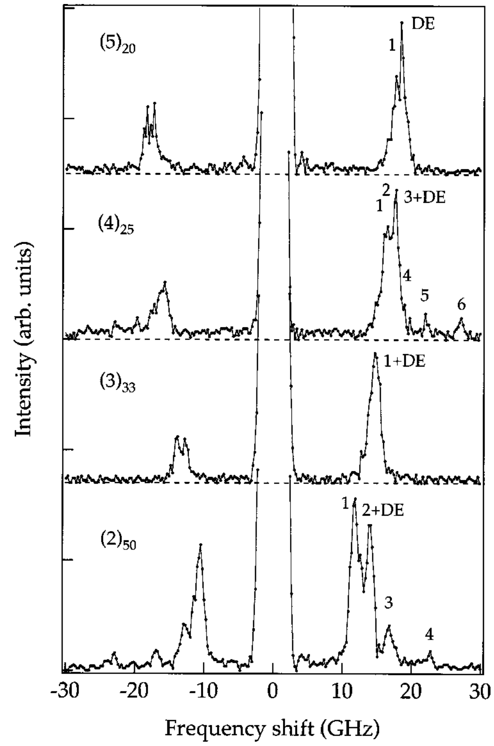

FIG. 2. A comparison of BS spectra observed from the integer-type SL's. The experimental conditions are the same as the ones given for Fig. 1. Total thickness of each SL is 34.5, 35.5, 35.5, and 36.4 nm from top to bottom. The labels 1 to 5 and DE stand for the SSW mode number and the DE mode.

trilayers it is already known that the IEC depends on a magnetic layer thickness[13] and also on cap layers added to the trilayers.[14] In our FLSL's these would mean that the coupling across one interlayer is not isolated at each interfaces, but there is a mutual interaction. Hence we cannot expect that the IEC values in these FLSL's are the same as in trilayers.

BS spectra were excited by the *p*-polarized 532 nm $(=\lambda)$/150 mW line from a solid state laser at 300 K. Backscattered beam was analyzed by using a six-pass tandem Fabry-Pérot interferometer[15] with a cross-polarized analyzer to eliminate scattering from surface acoustic phonons. Magnetic fields of up to 0.7 T were applied parallel to the film plane and perpendicular to the scattering plane. Spin waves (SW's) propagating along the crystallographic (110) direction in the Fe layer were examined. We also examined the surface dispersion (surface wave vector Q_S dependence of the SW frequencies under a constant magnetic field). Here, Q_S is defined by $Q_S = 4\pi \sin\theta/\lambda$ where θ is the incident angle measured from the surface normal. Typical spectrum accumulation time was around 2 h. The results are compared with *ab initio* predictions based on the self-consistent FLAPW method under the generalized gradient approximation (GGA). We report on IEC in $[\mathrm{Fe}(n\ \mathrm{ML})/\mathrm{Au}(n\ \mathrm{ML})]_m$ FLSL's with $n = 1$, 1.5, 1.75, 2, 2.25, 2.5, 2.75, 3, 3.25, 3.5, 4, and 5 and m the number of periods. We use for these FLSL's the abbreviation $(n)_m$.

Figure 1 displays BS spectra observed from $(n)_m$ FLSL's (where $n = 2 \pm \delta$, with $\delta = 0.25$ and 0.5). The spectra consist of a main SW peak (the strongest peak in each of the spectra) and additional features at higher frequencies, which are attributed to SSW as discussed already for the $(2)_{50}$ SL.[12] The SSW mode numbers are indicated on each spectrum. Except for the $(1.5)_{30}$ SL the frequency of the main peak increases with the SL period. This anomaly of the $(1.5)_{30}$ SL can be explained by the fact that with the available fields it was not possible to saturate this sample in the film plane, due to the extremely high perpendicular anisotropy. For the $(1.5)_{30}$ SL the SSW frequencies are larger than for the $(1.75)_{30}$ SL as clearly seen from the inserted spectra. The counterparts of these feature on the negative-frequency (Stokes) side are too weak to be observed. The frequencies of the SSW decrease with the SL period and are not clearly resolved in the case of the $(2.5)_{30}$ SL. Figure 2 shows a comparison of the BS spectra observed from the integer-type FLSL's with mode identification. The $(2)_{50}$ spectrum already shown in Fig. 1 is repeated for comparison. The trend of a frequency increase of the main line with the SL period still continues. Furthermore SSW features appear in the case of $(4)_{25}$ and $(2)_{50}$ but not for $(3)_{33}$ and $(5)_{20}$. This indicates that the interactions leading to the occurence of SSW are attenuated in the samples with $n = 3$, 5 as compared to the those with $n = 2$, 4. Furthermore the spectra from $(2)_{50}$ and $(3)_{33}$ can be smoothly connected by the two spectra with non-integer number of ML's at the bottom of Fig. 1.

For the assignment of the SW peaks of the odd-integer ML FLSL's, we examined the surface dispersion. Figure 3

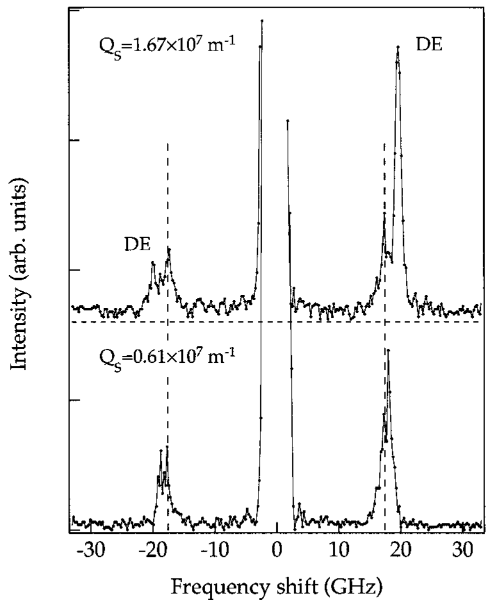

FIG. 3. Q_S development of SW spectra observed from the $(5)_{20}$ SL ($L_a=34.5$ nm) at $H=0.3$ T. The broken lines give the bulk SW frequencies of ± 17.6 GHz.

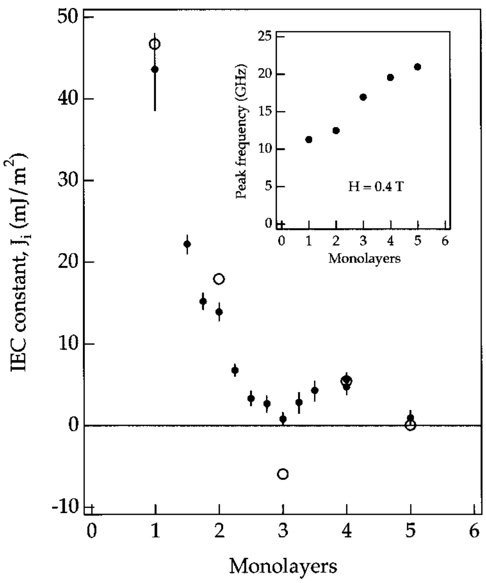

FIG. 4. The IEC constant J_i as a function of n; (●) BS and (○) *ab initio*. The inserted figure shows the main peak frequency of the bulk modes as a function of n obtained from the integer-type SL's at $H=0.4$ T.

displays BS spectra observed from $(5)_{20}$ for two values of Q_S. The frequency splitting of each doublet increases as the magnitude of Q_S increases. Although the lower-frequency peaks do not change their positions at ± 17.6 GHz as indicated by the broken lines, the higher-frequency peaks increase their frequencies and change their intensities. The anti-Stokes peak increases its intensity, but the Stokes peak decreases its intensity. Under the present experimental conditions, we expect the Damon-Eshbach (DE) surface localized SW on the laser illuminated surface on the anti-Stokes side. These frequency and intensity behaviors of the higher-frequency peaks have been widely observed for the DE mode in thin films.[16] The weak DE peak in the Stokes side is due to an amplitude leakage of the DE mode localized on the rear side of SL. The amplitude leakage is approximately given by a factor of $\exp(-Q_S L)$ with the film thickness of L. We confirmed similar surface dispersion effects for the $(3)_{33}$ SL. The singlet in the anti-Stokes side was actually an unresolved doublet. Hence, we assigned the lower-frequency peaks to the lowest-order SSW's and the higher-frequency peaks to the DE peaks for the odd-integer ML SL's.

SW frequencies were determined as a function of the magnetic field up to 0.7 T for all FLSL's. Because of the rapidly developing perpendicular magnetic anisotropy for shorter period SL's with $n<2$, our maximum field of 0.7 T was not strong enough to saturate the SL magnetization within the film plane. However, we could always observe SW scattering even for the unsaturated SL's. The inserted figure in Fig. 4 shows the lowest-order SSW frequency of the integer-type FLSL's as a function of n under a magnetic field of $H=0.4$ T. In order to analyze the SW frequencies as a function of the magnetic field, we regard the SL's as an ordered ''Fe-Au alloy'' film with perpendicular magnetic anisotropy and with an anisotropic exchange coupling.[12] Total thickness L_a of the ''alloy'' films was determined by XRD.[7] With this assumption, we can apply the conventional SSW approach to analyze the SW frequencies. The frequency of the nth SSW in a thin film, which satisfies the condition $(\pi/L_a)^2 \gg Q_S^2$, is given by

$$2\pi\nu(n) \cong \gamma[H+H_{ex}(n)]^{1/2}[H+H_{ex}(n)+4\pi M-H_A]^{1/2}, \tag{1}$$

where γ is the gyromagnetic ratio, $4\pi M$ the saturation magnetization of the alloy, H_A the perpendicular anisotropy field and $H_{ex}(n)$ the exchange field acting on the nth SSW. The exchange field is given by

$$H_{ex}(n)=D_{//}Q_S^2+D_\perp(n\pi/L_a)^2. \tag{2}$$

Here, $D_{//}$ and $D_\perp$ are the inplane and out-of-plane SW stiffness constants. $D_\perp$ is relevant for the SSW's and of interest here. We assumed a bulk Fe value of $D_{//}=2.3\times 10^{-17}$ Vm^2,[17] and introduced the diluted magnetization approximation $M=M_{Fe}d_{Fe}/(d_{Fe}+d_{Au})$ in which M_{Fe} is the magnetization of the Fe layer taking account of the enhancement of the magnetic moment per Fe atom, $2.65\pm 0.20\mu_B$ (Ref. 12) and d_{Fe} and d_{Au} are the thicknesses of Fe and Au layers determined by XRD.[7] Using Eqs. (1) and (2), we finally obtained a set of the magnetic constants, (γ, H_A, and $D_\perp$), for each SL. Then, we obtain the IEC strength $J_i=2MD_\perp/a$ (Ref. 18) where $a=0.287$ nm is the lattice constant of Fe. J_i is shown in Fig. 4 as a function of n. Although the IEC is always ferromagnetic, the $(3)_{33}$ SL gives the smallest value of $J_i\sim 0.8$ mJ/m^2. This is a typical value for the IEC constant measured at somewhat larger interlayer thickness.[19] The J_i value takes a maximum of ~ 5 mJ/m^2 for the $(4)_{25}$ SL, and then decreases to ~ 0.9 mJ/m^2 for the $(5)_{20}$ SL. As n decreases from 3, J_i rapidly increases and finally gives a value of $J_i=43.6\pm 5.0$ mJ/m^2 for the $(1)_{100}$ SL which corresponds to an ordered Fe-Au alloy film with the $L1_0$ structure. Since the $(1)_{100}$ SL has extremely strong perpendicular anisotropy, the maximum field of 0.7 T was too weak to pull down the magnetization into the film plane. Eventually, we have rather larger uncertainty for the J_i value. For pure Fe films in full contact the J_i value is expected to be 140 mJ/m^2.[18] Hence, the present value of $\sim$44 mJ/m^2 for the $(1)_{100}$ SL seems to be reasonable.

We also performed *ab initio* calculations for the integer-type FLSL's with $n=1$ to 5. The details of the calculation using the self-consistent FLAPW method under the GGA were described in Refs. 8–10. According to the *ab initio* results, *d* electrons from Fe atoms are almost isolated even by 1 ML of the Au interlayer. Then, the IEC is transmitted by itinerant *sp* electrons via second order processes which result in one order or more smaller J_i value compared with the direct one.[4,10,18] In Fig. 4, the *ab initio* values of J_i are plotted with open circles, which give fairly good agreements with the BS ones, except for $n=3$ where an antiferromagnetic (AF) ground state is predicted. Both of our SQUID and BS results indicate ferromagnetic ground state for $n=3$. As a possible orgin of the discrepancy between the experiments and calculation, we consider interface roughness. Since the IEC's for $n=2$ and 4 are ferromagnetic, the AF IEC for an ideal $n=3$ SL may be substantially smeared for long wavelength SW's seen by BS. We need more detailed studies to elucidate the even/odd behavior of J_i.

SW's in Fe(n ML)/Au(n ML) FLSL's with integer and noninteger ML ($n=1$ to 5) were examined by BS at 300 K. The IEC constant J_i has been evaluated as a function of n by applying the effective alloy model. The IEC is found to be ferromagnetic. We found a minimum of $\sim$0.8 mJ/m^2 at $n=3$ and a maximum of $\sim$5 mJ/m^2 at $n=4$. The J_i value rapidly increases as n decreases from 3 to 1, and finally becomes a value of $J_i=43.6\pm5.0$ mJ/m^2 for the $(1)_{100}$ SL which is an ordered Fe-Au alloy film with the $L1_0$ structure. We also performed *ab initio* calculations of J_i. The *ab initio* values of J_i give a good agreement with the BS values except for $n=3$, where an AF ground state is predicted. The even/odd effect on the IEC strength offers a quite new aspect of the IEC phenonemon, which is qualitatively different from the well-known oscillatory behavior of IEC observed in trilayers or superlattices consisting of a few nm or more thicker layers. However, the physical origin of the even/odd effect is not understood yet and left for future studies.

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