PHYSICAL REVIEW B VOLUME 34, NUMBER 12 15 DECEMBER 1986

Localized phonon modes in Fe-Pd multilayer structures

P. Baumgart, B. Hillebrands, R. Mock, and G. Güntherodt
II. Physikalisches Institut, Universität Köln, 5000 Köln 41, Federal Republic of Germany

A. Boufelfel and C. M. Falco
Department of Physics and Optical Sciences Center, University of Arizona, Tucson, Arizona 85721
(Received 9 July 1986)

The Rayleigh mode and up to six higher-order localized acoustic (Sezawa) surface modes have been observed in Fe-Pd multilayer structures using Brillouin scattering. The effective elastic constants of the multilayer structures have been determined from a fit of the measured mode dispersions. With decreasing multilayer modulation wavelength from 87 to 33 Å, the elastic constant c_{11} shows an interface-induced softening of up to 20%.

Currently there is considerable interest in the elastic and magnetic properties of metallic multilayer systems. However, the localized higher-order (Sezawa) phonon modes in these materials have so far not been investigated. Previous experimental work concentrated on only the Rayleigh surface mode.[1-3] While higher-order localized (Sezawa) phonon modes were observed in a few cases,[3,4] their quantitative analysis and use in a complete description of the elastic properties has been missing. Here we report the observation and measured dispersion of the higher-order localized phonon modes in Fe-Pd multilayer systems with 70–90 bilayers. The quantitative description, particularly of the higher-order localized phonon modes, yields the elastic constants of the multilayer structure and provides information about their modifications within a few angstroms of the interfaces due to induced strains. No dependence of the Rayleigh wave velocity on the modulation wavelength of the multilayer structures was found, contrary to results found in other systems.[1-3] The investigation of the spin-wave excitations of these multilayers is reported elsewhere.[5]

The Fe-Pd multilayer samples were prepared on epipolished sapphire substrates using a sputtering technique described elsewhere.[6] The sample parameters are listed in Table I. Bragg and wide-film Debye-Scherrer x-ray diffraction showed the layers grew with a preferred orientation of bcc Fe(110) planes and fcc Pd(111) planes with no correlation of the in-plane orientation among subsequent layers. Structural coherence extended at least 300 Å perpendicular to the layers. For each multilayer the number of atomic layers N_{at} in the Fe and Pd layers were equal. Hence the ratio d_1/d_2 of the thicknesses d_1 (d_2) of the Fe (Pd) layers is the same (=0.90) for all multilayer samples used here. The superlattice or modulation wavelengths L ($L = d_1 + d_2$) of the different samples were obtained from the x-ray diffraction spectra.

TABLE I. Parameters of the investigated Fe-Pd multilayer samples. N_{at} is the (equal) number of atomic layers in each single Fe and Pd layer.

Multilayer sample	Modulation wavelength L (Å)	Total bilayers N	N_{at}	Total thickness h (Å)
A	33.2	90	8	2988
B	40.1	90	9	3609
C	46.2	90	11	4158
D	64.6	70	15	4522
F	86.8	70	20	6076

The Brillouin scattering spectra were recorded at room temperature in backscattering geometry by means of a (3+3)-pass tandem Fabry-Perot interferometer.[7] p-polarized light of a single-moded 5145-Å Ar^+-ion laser with an incident power of up to 250 mW was focused on the samples, which were kept under vacuum. In order to avoid possible ambiguities caused by spin-wave excitations, a magnetic field of 3 kG was applied perpendicular to the scattering plane whereby the magnon peaks were shifted out of the phonon frequency range. A typical sampling time was $\frac{1}{2}$ h per spectrum.

Figure 1 shows a series of Brillouin spectra of the Fe-Pd multilayer samples (denoted A–F in order of increasing modulation wavelength, see Table I). The angle of incidence was 65°. The scattering intensities show the Stokes–anti-Stokes symmetry, which is characteristic of phonon excitations. The most intense inelastic peak in each spectrum at about 7 GHz frequency shift is due to the Rayleigh surface mode of the whole multilayer stack. At larger frequency shifts the higher-order (Sezawa) surface modes are observed.

Dividing the measured mode frequencies by the component $q_{\parallel}$ of the phonon wave vector parallel to the surface (which is determined by the scattering geometry) one obtains the phonon phase or sound velocities. The latter are plotted in Fig. 2 as a function of $q_{\parallel}h$, where h is the total thickness of the multilayer. Each symbol denotes a different sample as described in Fig. 2. A set of data points for the same sample was obtained by varying $q_{\parallel}$, i.e., by changing the angle of incidence. The choice of samples and of $q_{\parallel}$ allows the phonon velocities to be followed over a continuous range of $q_{\parallel}h$. A typical error bar is also indicated. The estimated mean error is ≈2% for $q_{\parallel}$, less than 1% for h, and ≈3% for the frequencies yielding a mean error of ±3.6% for v and ±2.2% for $q_{\parallel}h$.

© 1986 The American Physical Society

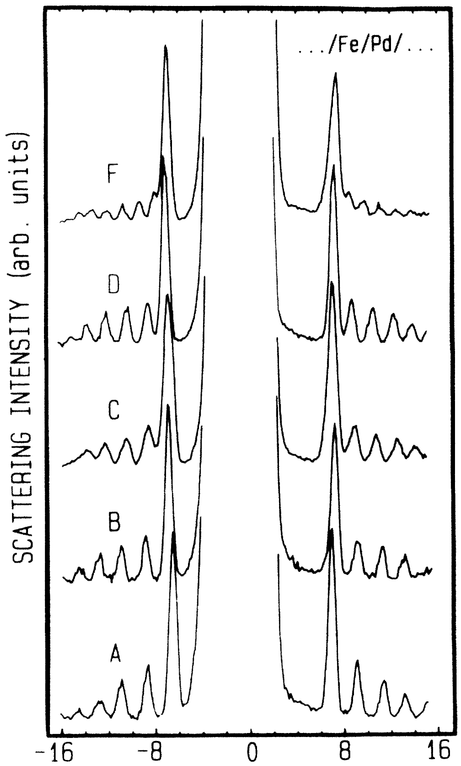

FIG. 1. Measured Brillouin spectra of Fe-Pd multilayer samples A–F (see Table I). The angle of incidence was 65°.

The sound velocities of the modes as a function of $q_{\parallel}h$ can be calculated by using a continuum mechanical model[8] and by taking into account appropriate boundary conditions at each interface.[9] However, this treatment requires a very high computational effort and therefore it cannot be applied to any fitting procedure. A more straightforward description is given by introducing effective elastic constants and by treating the multilayer stack as one homogeneous film of total thickness h.[10] This approach is justified in principle because the wavelengths of the phonon excitations under consideration are large compared to the multilayer modulation wavelength. However, the present work is the first experimental test of this concept. Besides the elastic constants of the two constituent materials, only the ratio d_1/d_2 enters the formulas for the effective elastic constants, which therefore should be the same for all Fe-Pd samples considered here. The sound velocities of the localized multilayer modes have been calculated by taking into account the appropriate mechanical boundary conditions only at the free surface of the film and at the substrate-film interface. It follows from this model that the sound velocities obtained are a function of the product $q_{\parallel}h$. To check the reliability of the concept of effective elastic constants the sound velocities of the modes of a ten bilayer stack on a substrate have also been calculated by the former explicit continuum mechanical method. For the considered range of $q_{\parallel}h$ $(0 \leq q_{\parallel}h \leq 15)$ we found the solutions to be indistinguishable from those obtained using the model of effective elastic constants (deviations smaller than 1%). For a proper assignment of the elastic constants we locate the interface in the x-y plane with phonon propagation in the x direction.

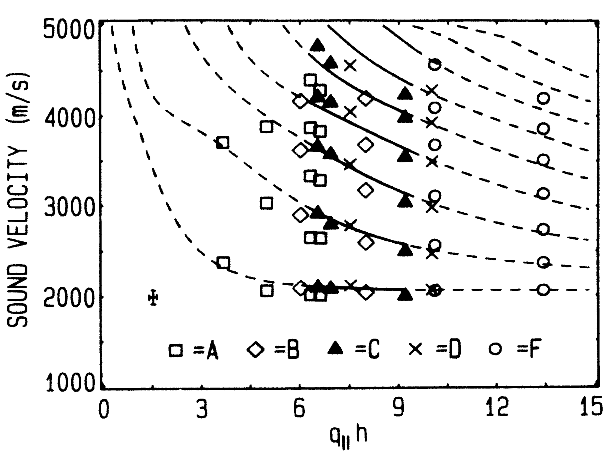

FIG. 2. Phonon sound (phase) velocities as a function of $q_{\parallel}h$. The symbols denote the values obtained for the different samples. The solid lines represent the theoretical fit with $c_{11}=225$ GPa for sample C (for the remaining parameters see text). The dashed lines are the extension of the fitted dispersion curves to the full range of $q_{\parallel}$h. A typical error bar is also indicated.

For the same order of localized phonon mode the measured sound velocities in Fig. 2 show discontinuities near the overlap of data points of subsequent samples. These discontinuities are larger for the higher-order modes. These experimentally observed discontinuities in the dispersion curves are in conflict with the above-discussed model. They can only be explained by assuming different effective elastic constants for the different multilayer samples as will be discussed in the following. Assuming the single layers to be isotropic leads to a cylindrical (or for the elastic tensor equivalently hexagonal) symmetry of the multilayer system. The elastic constants c_{11}, c_{13}, c_{33}, and c_{55}, but not c_{12}, enter in the calculation of the velocities of the Rayleigh and Sezawa modes. The effective elastic constants[10] calculated from the data of Ref. 11 for Fe and Pd are (in GPa) $c_{11}=271$, $c_{13}=135$, $c_{33}=273$, and $c_{55}=64.4$. These values together with the literature data for the sapphire substrate[11] (in GPa), $c_{11}=494$, $c_{12}=158$, $c_{13}=114$, $c_{33}=496$, $c_{44}=c_{55}=145$ (Ref. 12), were used as initial inputs for a fitting procedure[13] for the Rayleigh and Sezawa modes. From the effective elastic constants to be fitted, c_{55} is well decoupled from the rest since it determines mainly the Rayleigh mode for large $q_{\parallel}h$.[8] For all samples we found $c_{55} \approx 45.5 \pm 2$ GPa.[14] Thus, one major result of this present work is that no dependence of the Rayleigh velocity on the modulation wavelength of the multilayer samples was found, contrary to results found in other systems.[1-3] In addition to c_{55} a good quantitative fit for each multilayer sample can be achieved by fitting any one of the elastic constants c_{11}, c_{13}, or c_{33}, provided the other two are kept fixed at their starting values. We have taken c_{11} as the fitting parameter since the mismatch of approximately 10% between the lattice parameters of the Fe and Pd layers causes strains mainly parallel to the interfaces. Thus, we expect that elastic constants which correlate with stresses and strains parallel to the interfaces are most modified. The values obtained for the c_{11} range from 197 GPa for sample A to 243 GPa for sample F. The lines in Fig. 2 are calculated with $C_{11}=225$ GPa which is the fitted value of sample C. It can be seen that the data of this sample are fitted quite well[15] using this procedure. The dashed lines are the extension of these curves to the full range of $q_{\parallel}h$.

The variation of c_{11} found for the different samples leads to a model for the structure of the multilayer samples. Assume there is a constant thickness D of material at each interface, which has elastic constants different from the rest of the sample. The effective c_{11} is therefore composed of a modified c_{11}' within the fraction $2D/L$ at the interfaces and an unchanged c_{11}^0 for the rest of the bilayer,[10] i.e.,

$$c_{11}=\frac{2D}{L}c_{11}'+\left(1-\frac{2D}{L}\right)c_{11}^0 \ ,$$

yielding a linear dependence of c_{11} on $1/L$ for $2D<L$.[16] Figure 3 shows that the fitted values of c_{11} as a function of the inverse modulation wavelength give a straight line to a good approximation, in agreement with this model. Extrapolating this line to an infinite value of L actually reproduces the calculated starting value for c_{11} of 271 GPa. Using this model we can obtain an upper limit for the interface affected thickness D. Because in Fig. 3 the linearity extends at least down to $L=33$ Å, an upper limit to the thickness D in which c_{11} is changed can be estimated to be smaller than 16 Å per interface.

In summary, by measuring the Rayleigh mode only, no conclusions about elastic constants other than c_{55} can be drawn. The fitting procedure for the effective elastic constants described here shows that inclusion of the higher-order (Sezawa) surface modes allows a detailed investigation of the elastic properties of multilayer structures. In particular, information about interface-induced modifications of the elastic constants can be obtained, as can an upper limit for the interface thickness in which the elastic constants are modified. Since no broadening of the linewidths, especially of the higher-order Sezawa modes, could be detected, any roughness or island formation on the length scale of the wavelength of these phonons can be excluded. Thus the measurement and the analysis of the Sezawa modes of multilayer structures appear to be a powerful method to study their elastic behavior.

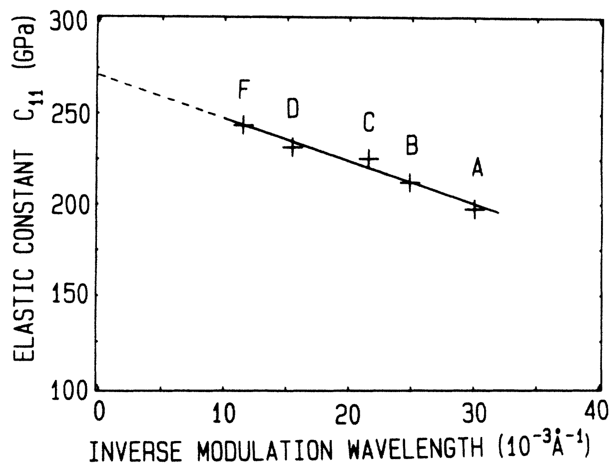

FIG. 3. Fitted values of the elastic constant c_{11} for the multilayer samples A–F as a function of their inverse modulation wavelength ($1/L$). The solid line is a linear least-squares fit through the measured points. The dashed extrapolation extends to large values of L, where the influence of a constant interface-affected thickness D (see text) can be neglected.

The work at Universität Köln was supported by Deutsche Forschungsgemeinschaft, Sonderforschungsbereich No. 125, and the work at the University of Arizona was supported by the U.S. Department of Energy, Materials Scineces Division under Contract No. DE-AC02-83ER45025.

[1]A. Kueny, M. Grimsditch, K. Miyano, I. Banerjee, C. M. Falco, and I. K. Schuller, Phys. Rev. Lett. **48**, 166 (1982); A. Kueny and M. Grimsditch, Phys. Rev. B **26**, 4699 (1982).

[2]M. R. Khan, C. S. L. Chun, G. P. Felcher, M. Grimsditch, A. Kueny, C. M. Falco, and I. K. Schuller, Phys. Rev. B **27**, 7186 (1983).

[3]A. Kueny, M. Grimsditch, B. Y. Jin, J. B. Ketterson, and J. B. Hilliard, J. Appl. Phys. **56**, 1550 (1984).

[4]G. Rupp, W. Wettling, W. Jantz, and R. Krishnan, Appl. Phys. A **37**, 73 (1985).

[5]B. Hillebrands, P. Baumgart, R. Mock, G. Güntherodt, A. Boufelfel, and C. M. Falco, preceding paper, Phys. Rev. B **34**, XXX (1986).

[6]C. M. Falco, J. Phys. (Paris) Colloq. **45**, C5-499 (1984).

[7]R. Mock, B. Hillebrands, and J. R. Sandercock (unpublished).

[8]G. W. Farnell, in *Physical Acoustics*, edited by W. P. Mason and R. N. Thurston (Academic, New York, 1972).

[9]B. Hillebrands, Ph.D. thesis, University of Köln, 1986 (unpublished).

[10]M. Grimsditch, Phys. Rev. B **31**, 6818 (1985).

[11]*Elastic, Piezolelectric, Pyroelectric, Piezooptic, Electrooptic Constants, and Nonlinear Dielectric Susceptibilities of Crystals*, Landolt Börnstein, New Series, Group 3, Vol. 18, edited by K.-H. Hellwege and A. M. Hellwege (Springer-Verlag, Berlin, 1984).

[12]For the sake of simplicity, the small deviation of the crystalline structure of sapphire from othorhombic symmetry has been neglected, i.e., the value of $c_{14}=-23$ GPa has been set equal to 0.

[13]B. Hillebrands, P. Baumgart, R. Mock, and G. Güntherodt, J. Appl. Phys. **58**, 3166 (1985).

[14]The multilayer samples were found experimentally to be isotropic in the planes parallel to the interfaces. Because no method for averaging the elastic constants in only two dimensions is

known, the single layers were assumed to be isotropic in all direction, neglecting the preferred orientations of growth. This method gives the starting value $c_{55} = 64.4$ GPa. On the other hand, neglecting this two-dimensional isotropy and considering the Fe (Pd) layers oriented in [110] ([111]) directions parallel to the z axis gives a value of $c_{55} = 51 \pm 7$ GPa, where the range of values is determined by the in-plane direction of phonon propagation. This value is quite close to that found by the fit.

[15]Using the fitted values of c_{11} which are different for each sample would give a discontinuous and thereby very puzzling plot of the dispersion curves for the higher-order modes in Fig. 2.

[16]The cross terms in Ref. 10 have been omitted, because they are quite small (about 5%).