Polarization effects and interlayer coupling in (Fe-Si)/Pd multilayered films

1992 J. Phys.: Condens. Matter 4 9893

(http://iopscience.iop.org/0953-8984/4/49/016)

J. Phys.: Condens. Matter 4 (1992) 9893–9900. Printed in the UK

Polarization effects and interlayer coupling in (Fe–Si)/Pd multilayered films

Yi-Hua Liu†‡, Xiao-Ding Ma‡ and Liang-Mo Mei‡

† Chinese Centre of Advanced Science and Technology (World Laboratory), PO Box 8730, Beijing 100080, People's Republic of China
‡ Department of Physics, Shandong University, Jinan, Shandong 250100, People's Republic of China

Received 24 April 1992, in final form 6 July 1992

Abstract. (Fe–Si)/Pd multilayered films with different layer thicknesses have been obtained by RF sputtering. When the thickness of the Pd layers is fixed, the specific saturation magnetization M_s of films increases with decrease in the thickness of Fe–Si layers. This result is caused by the polarization effect of Pd atoms. On varying the thickness d_p of the Pd layers, with a fixed Fe–Si layer thickness, it is found that for Pd layers thinner than 36 Å the polarization effect of Pd is reduced and even disappears at $d_p = 18$ Å; then magnetic coupling between the magnetic layers appears on further decrease in d_p.

1. Introduction

Recently, interest in Pd in the study of multilayered structures has increased strongly. Many experiments have shown that magnetization enhancement exists in multilayered films consisting of magnetic layers and Pd metal layers [1–3]. This enhancement is induced by the polarization effect of Pd atoms. A free Pd atom is non-magnetic. However, the intra-atomic exchange interactions between 4d electrons are important, and an enhanced magnetic susceptibility of the Pd metal can result. Additional ferromagnetism can be induced in Pd layers by the magnetic atoms through the conductive electrons. This is a kind of RKKY-like exchange interaction. Recently, experimental and theoretical results showed that for some normally non-magnetic transition metals, such as Rh or Pd, the onset of ferromagnetism can occur for expanded volumes, and their magnetic moments increase with increasing volume with an infinite slope [3–5]. In Fe/Pd/Fe trilayers [3, 5], the laterally stretched Pd spacer had ferromagnetic order throughout the whole Pd layer when the thickness of the Pd layer was four monolayers. In this case, Pd couples Fe layers strongly in a ferromagnetic manner.

When the non-magnetic layers in the multilayered films are sufficiently thin, magnetic coupling between magnetic layers begins to occur. It was found that ferromagnetic or antiferromagnetic coupling can exist between magnetic layers separated by Cu, Cr, Pd or Ru layers, and an oscillatory interlayer coupling was found with a variation in the thickness of non-magnetic layers [6, 7]. Some theoretical attempts have been made to identify the origin of this coupling [8, 9]. When the interlayer coupling changes, the magnetization orientation in adjacent magnetic layers

0953-8984/92/499893+08$07.50

will change from parallel to antiparallel, or the reverse. Investigation of the magnetic coupling between magnetic layers is also important in the study of multilayered films. In this work, we used magnetic measurements to study the polarization effect of Pd and the coupling between magnetic layers in (Fe–Si)/Pd multilayered films. Fe–Si alloy can be easily fabricated in the amorphous state, making it easier to match it with non-magnetic layers in the multilayered structures, and Fe–Si amorphous alloy has been extensively studied [10–12]. One can use this multilayered structure to investigate the magnetic interactions between Fe–Si amorphous alloy and Pd metal layers. Many studies of the multilayered films consisting of Pd and transition metals have been reported, but few deal with multilayered films consisting of Pd layers and amorphous magnetic layers.

2. Experimental details

Specimens were prepared with a RF sputtering system that consists of two targets. The targets were discs made of Fe–Si alloy and Pd. The distance between the target and substrate holder, each of which was water cooled, was about 4 cm. The RF input power was about 180 W. After the chamber had been evacuated to a high vacuum of about 2×10^{-6} Torr, Ar gas 99.999% pure was introduced. During the sputtering process, the Ar charging pressure in the chamber was kept at 5×10^{-3} Torr. The substrates were glass slides of 0.2 mm thickness. Single-layer Fe–Si films with soft magnetic properties were obtained with this system. The composition of the Fe–Si films determined by electron microprobe analysis was $Fe_{80.5}Si_{19.5}$. This is the lower limit of the composition where the amorphous nature can be observed [10]. (Fe–Si)/Pd multilayered films with different modulation wavelengths were made by sputtering Fe–Si and Pd on the substrate alternately and controlling the sputtering time according to the deposition rates. The rates were 0.85 Å s^{-1} and 2.83 Å s^{-1} for Fe–Si and Pd, respectively. Two series of samples were obtained with this system. Their thicknesses are shown in table 1. The total number of bilayers was 40 for all samples. X-ray diffraction measurements showed that crystalline peaks were not found for single-layer Fe–Si films nor for the second series of (Fe–Si)/Pd samples with Pd layers thinner than 36 Å. For the first series of samples and the second series with thicker Pd layers, x-ray diffraction showed that only a small broadened Pd(111) diffraction peak could be seen, but no crystalline peak could be found for Fe–Si layers. So Fe–Si layers in all samples are in an amorphous state, and Pd layers are in a microcrystalline state or perhaps an amorphous state for some samples with thinner Pd layers.

Table 1. Fe–Si layer thicknesses d_m and Pd layer thicknesses d_p for two series of (Fe–Si)/Pd multilayered films.

Series 1	d_m (Å) (fixed $d_p = 54$ Å)	7.5	15	21	30.6	40	48.5	80	89.3	120
Series 2	d_p (Å) (fixed $d_m = 15$ Å)	10.8	14.4	18	25.2	36	54	72	126	

The magnetic properties of (Fe–Si)/Pd films have been measured with a microprocessor controlling a vibrating-sample magnetometer. The specimens used in the measurements had an area of 5 mm × 8 mm. The applied magnetic field was in the plane of the films. The temperature dependences of the magnetic properties of samples were measured using two different systems. A Dewar cryostat was used for the low-temperature region from 77 to 300 K with the temperature varied at a rate of about 7 K min^{-1}, and an oven was used for the high-temperature region from room temperature to 900 K with the temperature varied at a rate of about 12 K min^{-1}. The sample in the oven was in a vacuum of 10^{-3} Torr in order to protect the sample against oxidation. The magnetic background signals of the substrate and the sample holder were considered. Their influences were small to negligible except for the samples with a weak magnetic signal.

3. Results and discussion

The temperature dependences of the specific saturation magnetization M_s for the first series of samples were measured in the temperature region from 77 to 700 K. The applied magnetic field used in the measurements was 500 Oe, which was high enough to saturate magnetically all samples because of their low coercive forces as shown from the typical loops in figure 1. The loop in figure 1(*d*) was obtained at 77 K; the others were obtained at room temperature. Figure 1(*a*) is a loop with a maximum coercive force at room temperature in our measurements; it indicates that the applied field used here was high enough to make samples saturate. Figure 2 shows some typical M_s versus T curves obtained from the first series of samples. The broken lines below 77 K are obtained by extrapolating curves according to the Bloch $T^{3/2}$ law. The experiments showed that the temperature dependence of the saturation magnetization at low temperatures for the multilayered films obeys this law [13, 14]. We do not show other M_s versus T curves because they are so close to each other. Figure 3 shows the M_s versus T curves obtained from the second series of samples. The measurement conditions were the same as those used in figure 2.

Curves A and B in figure 4 show the dependences of M_s at 0 K and room temperature, respectively, on the thickness d_m of Fe–Si layers obtained from figure 2. The full circles in figure 4 are the experimental data. One can see from figure 4 that M_s increases monotonically with decreasing d_m and is always larger than that of single-layer Fe–Si films (see figure 5). The reason for the increase in M_s for (Fe–Si)/Pd films is the polarization effect of Pd atoms, as indicated in many experiments [1–3]. When d_m decreases, the influence of the Pd polarization effect increases relatively; so the average M_s of the samples increases too. One can assume that the magnetic enhancement of a multilayered film caused by the polarization of Pd atoms is equivalent to an increment Δd of the Fe–Si layer thickness. Then a relation between M_s and d_m can be obtained as follows:

$$M_s(d_m) = M_s(\infty)(1 + \Delta d/d_m) \tag{1}$$

where $M_s(\infty)$ and $M_s(d_m)$ are the specific saturation magnetizations of amorphous Fe–Si bulk alloy and (Fe–Si)/Pd films with a thickness d_m of the Fe–Si layers. The results fitted according to equation (1) are shown by the full curves A and B in figure 4, where the data used for the fit are as follows: $M_s(\infty) = 196$ emu g^{-1} and $\Delta d = 6.3$ Å at 0 K; $M_s(\infty) = 173$ emu g^{-1} and $\Delta d = 2.7$ Å at room temperature.

The polarization effect of Pd atoms occurs mainly at the interfaces. Fe atoms at the interfaces induce neighbouring Pd atoms to polarize through the conductive

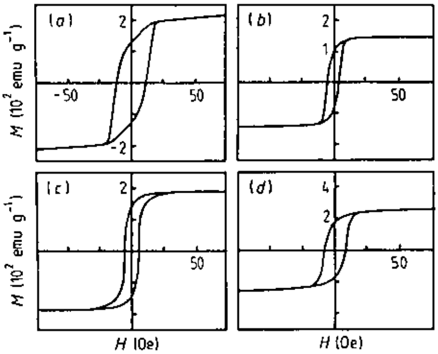

Figure 1. Typical loops of (Fe–Si)/Pd multilayered films (d_m = 15 Å): (*a*) d_p = 10.8 Å, room temperature; (*b*) d_p = 18 Å, room temperature; (*c*) d_p = 126 Å, room temperature; (*d*) d_p = 36 Å, 77 K.

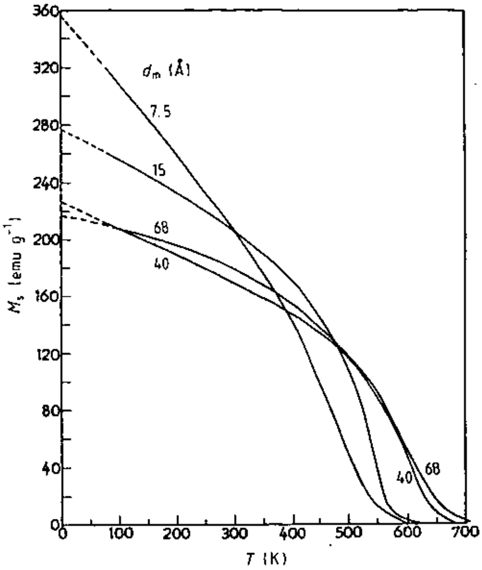

Figure 2. Specific saturation magnetization M_s versus temperature T for (Fe–Si)/Pd multilayered films. The data on the curves indicate the Fe–Si layer thickness d_m (Pd layer thickness d_p = 54 Å).

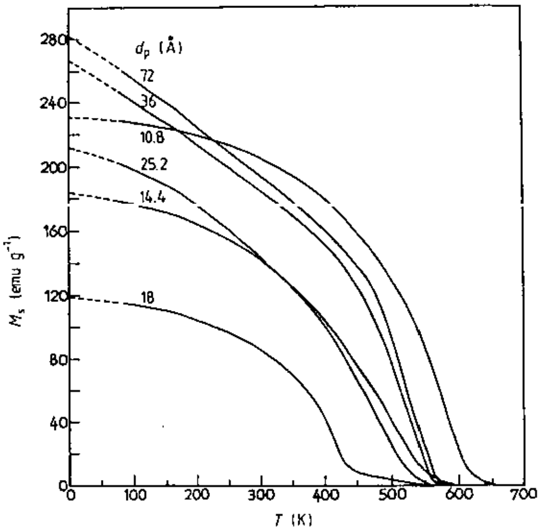

Figure 3. Specific saturation magnetization M_s versus temperature T. The data on the curves indicate the Pd layer thickness d_p (Fe–Si layer thickness d_m = 15 Å).

electrons via the intra-atomic exchange interactions between the band electrons of Pd atoms. So an additional magnetic moment results. There is a range of concentrations of Fe atoms at the interfaces because of atomic interdiffusion during the sputtering process. The transition temperature T_C of Pd polarization must be distributed over a wide range because T_C is very sensitive to the concentration [15]. This causes

M_s to decrease rapidly with increasing temperature at low temperatures as shown in figure 2, especially for the samples with thinner Fe–Si layers. At high temperatures, the atomic interdiffusion becomes important, and the alloying process destroys the multilayered structure. The Curie temperature of Fe–Pd–Si alloys is lower than that of Fe–Si alloys; so the magnetic properties of Fe–Si alloys at high temperatures cannot be detected in figures 2 and 3. Figure 5 plots the temperature dependences of M_s for a single-layer Fe–Si film (d_m = 1800 Å) and for a multilayered (Fe–Si)/Pd film with the thickest Fe–Si layers of 120 Å (d_p = 54 Å). In this case, the characteristics of remnant Fe–Si layers are exposed. For the single-layer Fe–Si film, a crystallizing transition at around T = 500 K and a phase transition of crystalline Fe–Si alloy at around T = 660 K can be seen [16]. For (Fe–Si)/Pd films, one can see two transition points on the M_s versus T curve. At T = 630 K, the alloying process of Fe, Si and Pd is no longer important. The magnetic properties of remnant Fe–Si layers begin to appear. M_s for remnant Fe–Si layers decreases more slowly than that of Fe–Pd–Si alloys, as can be seen from the curves for $T \geqslant 630$ K. So the phase transition of the crystalline Fe–Si alloy at around T = 660 K can also be seen in this case. One can see from figures 2 and 3 that the Curie temperatures T_C of Fe–Pd–Si alloys range from 530 to 670 K. The thinner the Fe–Si layers, the lower is T_C because of the higher Pd content in the Fe–Pd–Si alloys.

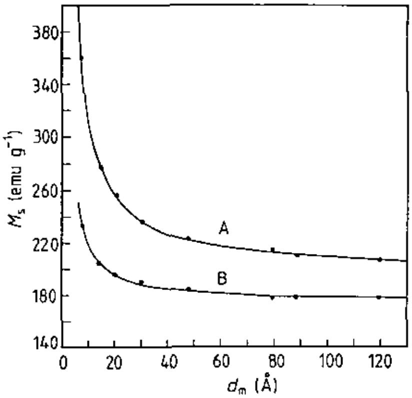

Figure 4. Dependences of M_s on d_m obtained from extrapolating the value of M_s to 0 K (curve A) and obtained at room temperature (curve B): ——, results from the fitting according to equation (1).

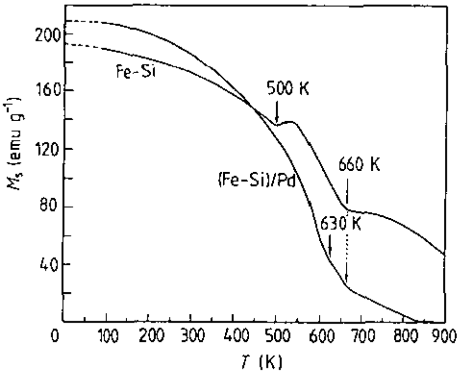

Figure 5. Specific saturation magnetization M_s versus temperature T for a single-layer Fe–Si film (1800 Å thickness) and an (Fe–Si)/Pd multilayered film with d_m = 120 Å and d_p = 54 Å. The arrows on the curves indicate the transition temperatures.

Curves A and B in figure 6 show the dependences of M_s at 0 K and room temperature on the thickness d_p of Pd layers obtained from figure 3. When d_p is smaller than 36 Å, M_s decreases rapidly with decreasing d_p and reaches a minimum at d_p = 18 Å. As d_p decreases further, M_s increases sharply. It seems that the polarization effect of Pd atoms decreases with decreasing d_p from d_p = 36 Å and disappears when d_p = 18 Å. When the polarization effect of Pd atoms disappears, i.e. when the magnetic interactions between Pd and Fe atoms at the interfaces disappear, a dead layer can be formed at the interfaces of Fe–Si layers, as occurs in such multilayers as (Fe–Si)/Si [16], Ni/Cu [17] and Fe/Bi [18]. In order to understand the reduction in magnetization, the conversion electron Mössbauer spectra of the samples were recorded at room temperature. Figure 7 shows three of these spectra, in which the full curves are the theoretical fits (a more detailed conversion electron Mössbauer spectroscopy analysis for all samples will be published elsewhere). Figure 7(*a*) is a spectrum of a single-layer Fe–Si film for comparison; this is a typical amorphous spectrum with broadened lines. When $d_p > 36$ Å, the shape of spectra for (Fe–Si)/Pd films is similar to that in figure 7(*a*). When $d_p = 18$ Å, a strong paramagnetic component in the spectrum appears, as shown in figure 7(*b*). It is similar to the case observed for (Fe–Si)/Si films with thinner Fe–Si layers [16]. This paramagnetic part of the spectrum originates from the non-magnetic part of the interfaces of multilayers, i.e. the dead layers.

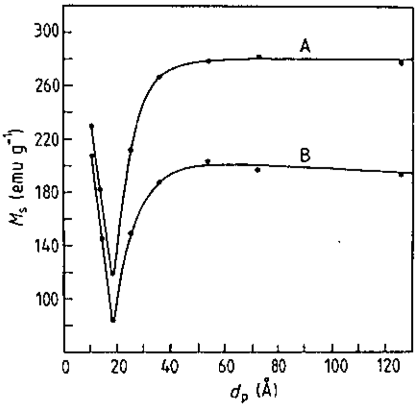

Figure 6. Dependences of M_s on d_p obtained from extrapolating the value of M_s to 0 K (curve A) and obtained at room temperature (curve B).

The thickness d_D of the effective dead layers can be determined from the following equation [17]:

$$M_s(d_m) = M_s(\infty)(1 - d_D/d_m) \tag{2}$$

where $d_m = 15$ Å. Using the results of fitting $M_s(\infty)$ in figure 4, and the experimental value of $M_s(d_m)$ at $d_p = 18$ Å, d_D can be obtained: $d_D = 5.9$ Å at 0 K and $d_D = 7.8$ Å at room temperature. In the study on (Fe–Si)/Si multilayered films we found [16] that the thickness of a dead layer in (Fe–Si)/Si films is 5.8 Å at 0 K, and 8.1 Å at room temperature. The results obtained from the two cases are almost the same. The reason that the polarization effect of Pd atoms disappears is unclear as yet. It is perhaps related to a change in microstructure of Pd layers such as a transition from the crystalline to the amorphous state.

When d_p decreases further from $d_p = 18$ Å, M_s increases rapidly. One can see from figure 3 that, when $d_p \leqslant 18$ Å, the shapes of the M_s versus T curves below room temperature are quite different from those for $d_p > 18$ Å. The curves at low temperatures are much flatter than those for $d_p > 18$ Å. This implies that the enhancement mechanism of M_s at small d_p is different from that at larger d_p, i.e. the polarization of Pd atoms will perhaps not be dominant at small d_p. In this case, exchange coupling between Fe–Si layers appears. It was considered that the

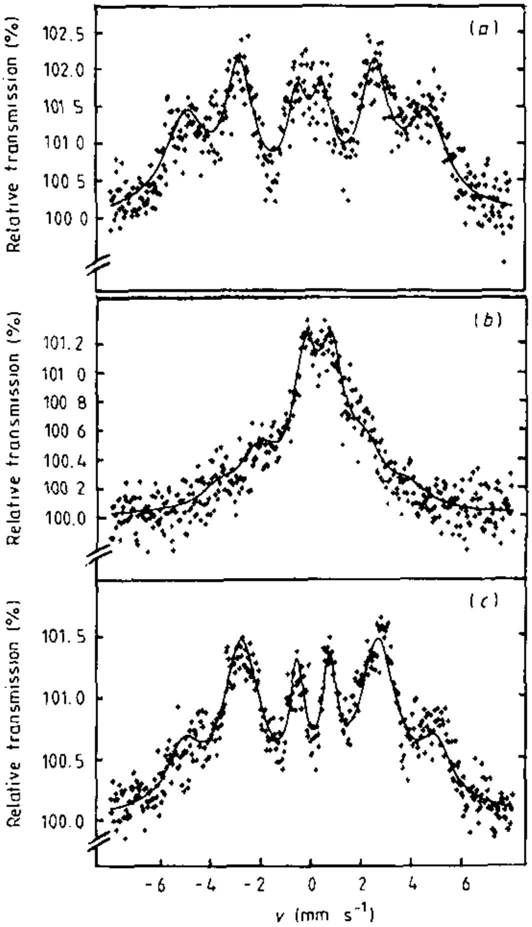

Figure 7. Conversion electron Mössbauer spectra at room temperature for (*a*) a single-layer Fe–Si film (thickness, 1800 Å), (*b*) an (Fe–Si)/Pd film with a Pd layer thickness $d_p = 18$ Å ($d_m = 15$ Å) and (*c*) an (Fe–Si)/Pd film with $d_p = 10.8$ Å ($d_m = 15$ Å).

Fe atoms which diffused into the Pd layers are responsible for this coupling. This is similar to what happens in the (Fe–Si)/Si multilayered films [16]. When the Pd layers are sufficiently thin, the Fe atoms which diffused into the Pd layers from the Fe–Si layers of both sides of the Pd layers intermix. The paramagnetic Fe atoms residing in the dead layers at the interfaces magnetically interact through the Fe atoms in the Pd layers [19]. This reduces the number of paramagnetic Fe atoms at the interfaces. The thickness of the effective dead layers decreases accordingly, and the magnetism of the films is strengthened. The conversion electron Mössbauer spectrum shown in figure 7(*c*) confirms that the interlayer coupling has obviously reduced the paramagnetic component of the Mössbauer spectrum.

4. Conclusions

The magnetic properties of (Fe–Si)/Pd multilayered films have been studied in detail. An obvious enhancement of magnetization has been observed when the Pd layers are thick. This is caused by the polarization of Pd atoms. The strength of polarization is temperature dependent and independent of the thickness of magnetic layers. When the thickness of the Pd layers decreases to a critical value, the polarization of Pd atoms is reduced and even disappears at a well defined thickness of Pd layers. On further decrease in the Pd layer thickness, an exchange-coupling effect between the magnetic layers appears. This causes the magnetism of films to strengthen again.

Acknowledgments

This work was supported by the National Science Foundation of China, the Doctoral Training Foundation of the National Education Commission and the State Key Laboratory of Magnetism, Institute of Physics, Chinese Academy of Sciences.

References

[1] Schröder K 1985 *J. Appl. Phys.* **57** 3666

[2] den Broeder F J A, Donkersloot H C, Draaisma H J G and de Jonge W J M 1987 *J. Appl. Phys.* **61** 4317

[3] Heinrich B, Celinski Z, Myrtle K, Cochran J F, Arrott A S and Kirschner J 1991 *J. Magn. Magn. Mater.* **93** 75

[4] Moruzzi V L and Marcus P M 1989 *Phys. Rev.* B **39** 471

[5] Celinski Z, Heinrich B, Cochran J F, Muir W B, Arrott A S and Kirschner J 1990 *Phys. Rev. Lett.* **65** 1156

[6] Celinski Z and Heinrich B 1991 *J. Magn. Magn. Mater.* **99** L25

[7] Parkin S S P, More N and Roche K P 1990 *Phys. Rev. Lett.* **64** 2304

[8] Levy P M, Ounadjela K, Zhang S, Wang Y, Sommers C B and Fert A 1990 *J. Appl. Phys.* **67** 5914

[9] Wang Y, Levy P M and Fry J L 1991 *J. Magn. Magn. Mater.* **93** 395

[10] Shimada Y and Kojima H 1976 *J. Appl. Phys.* **47** 4156

[11] Kobliska R J, Aboaf J A, Gangulee A, Cuomo J J and Klokholm E 1978 *Appl. Phys. Lett.* **33** 473

[12] Tsunashima S, Mitsuya S and Uchiyama S U 1979 *Japan. J. Appl. Phys.* **18** 1645

[13] Xiao J Q, Gavrin A, Xiao G, Childress J R, Bryden W A, Chien C L and Edelstein A S 1990 *J. Appl. Phys.* **67** 5388

[14] Martinez B, Moreu M A, Labarta A, Obradors X and Tejada J 1988 *J. Appl. Phys.* **63** 3206

[15] Wohlfarth E P (ed) 1980 *Ferromagnetic Materials* vol 1 (Amsterdam: North-Holland) p 155

[16] Liu Y H, Ma X D and Mei L M 1992 *Phys. Rev.* B **45** 10459

[17] Xiao G and Chien C L 1987 *J. Appl. Phys.* **61** 4061

[18] Cui F Z, Fan Y D, Wang Y, Vredenberg A M, Draaisma H J G and Xu R 1990 *J. Appl. Phys.* **68** 701

[19] Kazama N S and Fujimori H 1983 *J. Magn. Magn. Mater.* **35** 86