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MAGNETO-OPTICAL CHARACTERIZATION OF FeAu ARTIFICIALLY ORDERED ALLOY

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Abstract: Superlattices consisting of Fe and Au ultrathin layers with thickness of a few monolayers were prepared. Formation of $L1_0$-type structure in the Fe(1ML)/Au(1ML) superlattice was confirmed by X-ray diffraction. Magneto-optical studies suggest formation of a new energy band structure characteristic of the new alloy.
KEYWORDS: Fe/Au multilayers, artificially ordered alloy, magneto-optical polar Kerr effect, spectrum

INTRODUCTION

Recent development of epitaxial growth technology has made it possible to fabricate artificial structures on an atomic scale.[1] In particular, artificial structures (superlattices or sandwiches) consisting of magnetic and nonmagnetic thin layers have been attracting interest as new functional materials with novel physical properties such as giant magneto-resistance (GMR), large surface magnetic anisotropy and peculiar magneto-optical response characteristic of quantum confinement.[2]

In most of the studies, the physical properties of the layers in the artificial structures, no matter whether they are magnetic or nonmagnetic, have been presumed to keep the same crystal and electronic structures in multilayers as those in the bulk. Only a few investigations have been carried out from the view point that the artificial structure shows completely different electronic structure from those of its constituent layers in their free-standing states.

Recently we succeeded in the artificial preparation of a novel ordered alloy with $L1_0$ crystal structure by means of alternate deposition of Fe and Au layers with monoatomic thickness on a Au (001) buffer layer grown on a MgO (001) substrate.[3,4] Atomic arrangement in a unit cell of the tetragonal $L1_0$ ordered structure is given in Fig. 1. Fabrication of such "ordered alloy" structure is quite remarkable, since the Fe-Au system is known to show a peritectic-type phase diagram, which precludes existence of neither intermetallics nor intermediate alloys.

In order to clarify whether the electronic structure of the artificially ordered alloy is that expected from a simple stack of two kinds of material or a completely new one, we measured magneto-optical spectra on the artificial alloy. We found that the magneto-optical spectrum of the artificial Fe-Au alloy shows a distinguishing feature which is not found in a Fe film or in a Fe/Au multilayer with thick layers.[5]

In the present study we prepared superlattices consisting of N periods of n monoatomic layers (ML) of Fe and Au (hereafter denoted as $[Fe(nML)/Au(nML)]_N$) on Au (001)/MgO (001) as well as on Au (111)/Al_2O_3 (11.0) substrates and measured magneto-optical Kerr spectra in these films.

EXPERIMENTAL

The specimens used in the present study have been prepared by an ultra high vacuum (UHV) deposition technique on either MgO (100) or Al_2O_3 (11·0) substrates. The base pressure of the deposition system was 3×10^{-10} Torr.

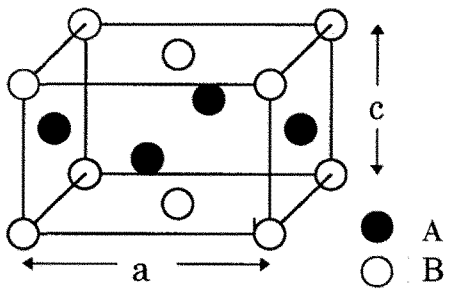

Fig.1 Tetragonal $L1_0$ ordered structure

A Fe seed layer of 1 nm followed by a Au buffer layer of 50 nm was deposited at 200°C and subsequently annealed for 30 min to 1 h at 500 °C. The orientation of the Au buffer layer was (001) for MgO substrates and (111) for Al_2O_3. The Fe seed was necessary to control the orientation of the Au layer. Multilayers with N periods, each of which consists of n ML Fe layers and n ML Au layers, were deposited in the UHV system at 70 °C on the Au buffer. The orientation of the film surface in Fe on Au(100) was (001), while that on Au(111) was (110). The deposition rates were approximately 0.01 nm/s. The layer thickness was controlled by using a quartz thickness monitor. Details of preparation techniques were described elsewhere.[3,4,6] Epitaxial growth of Fe/Au multilayers were confirmed by RHEED patterns for both series of samples.

These films were characterized by X-ray diffraction (XRD). The $[Fe(1ML)/Au(1ML)]_{100}$ multi-layer on Au(001) clearly showed a diffraction line (2θ=22.9°) characteristic of the tetrahedral $L1_0$ ordered structure. For Fe/Au multilayers with thicker layers, i.e., $[Fe(n ML)/Au(n ML)]_N$ (n=2-5), satellite lines due to multilayered structure were clearly observed.

On the other hand, no such diffraction lines were observed in the $[Fe(1ML)/Au(1ML)]_{100}$ film on Au (111). This fact rules out formation of any ordered structure in this multilayer. However, in $[Fe(n ML)/Au(n ML)]_N$ (n=2 and 3) on Au(111), well-defined superlattice lines of 1st, 3rd and 5th order were observed, indicating the formation of a superstructure.

Table I Structure parameters of $[Fe(n ML)/Au(n ML)]_N$ multilayers

Au buffer orientation	n (ML)	N (periods)	thickness for one period (nm)	average spacing (nm)
Au(001)	1	100	0.3800	0.190
	2	50	0.7272	0.1818
	3	33	1.0744	0.1791
	4	25	1.4216	0.1777
	5	20	1.7688	0.1769
	10	10	5.600	0.2800
Au(111)	1	100	0.448	0.224
	2	50	0.892	0.223
	3	33	1.344	0.224

The designed layer thickness, the number of periods and the average lattice-plane spacing normal to the film plane obtained by XRD are listed in Table 1.

Magneto-optical Kerr rotation and ellipticity were measured at room temperature between 1.2 and 6 eV using a specially designed Kerr spectrometer.[7] The maximum field applied was 1.7 T. Optical reflectivity was measured using a Hitachi U-3410 spectrophotometer, from which optical constants were deduced by the Kramers-Kronig analysis with the help of the data values measured by a Woollam type WVASE spectroscopic ellipsometer between 2 and 5 eV.

RESULTS AND DISCUSSION

Fe/Au MULTILAYERS ON Au(001)

Figure 2 illustrates spectra of polar magneto-optical Kerr rotation θ_K (solid line) and Kerr ellipticity η_K (dotted line) in the $[Fe(10ML)/Au(10ML)]_{10}$ multilayer on Au(001) buffer, which shows a prominent peak of Kerr rotation θ_K and a well-defined dispersion-type Kerr ellipticity η_K at 2.4 eV. This structure may be caused by the enhancement of MO signal by rapid change of optical constants in the Au layers as simulated by means of virtual optical constant method described in the preceding paper.[5]

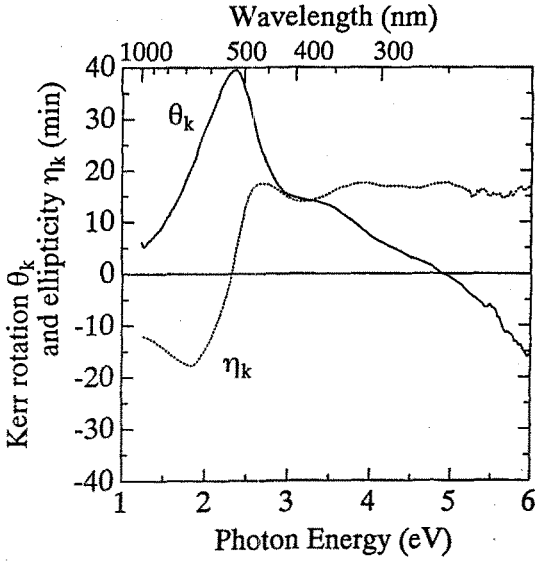

Fig. 2 Spectra of magneto-optical Kerr rotation θ_K (solid curve) and Kerr ellipticity η_K (dotted curve) in $[Fe(10ML)/Au(10ML)]_{10}$ multilayers on Au(001) buffer

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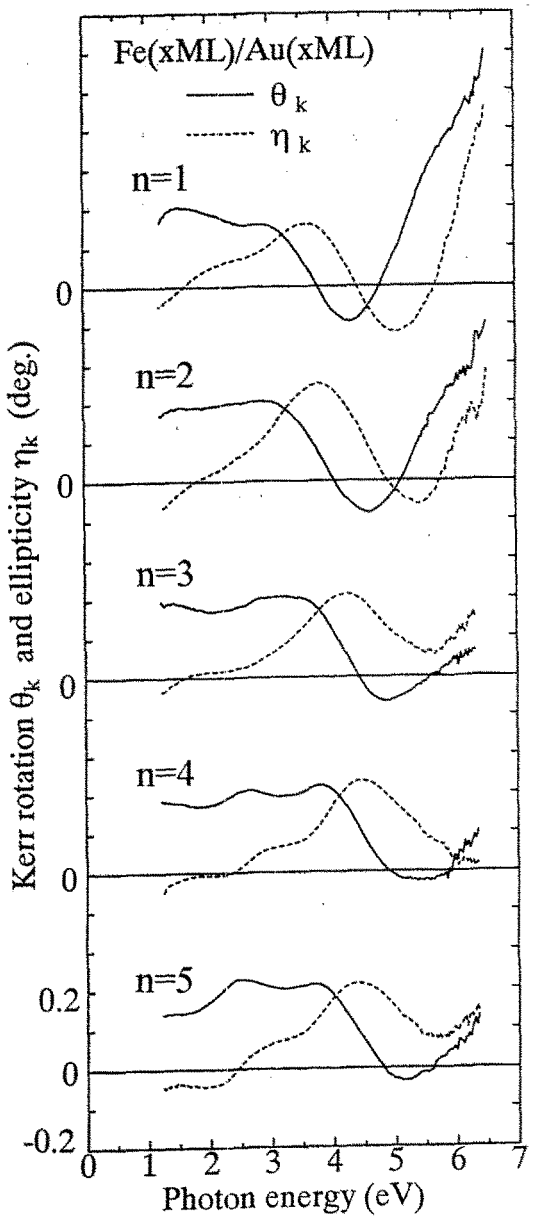

Fig. 3 Spectra of magneto-optical Kerr rotation θ_K (solid curve) and Kerr ellipticity η_K (dotted curve) in Fe(nML)/Au(nML) ($1 \leq n \leq 5$) multilayers on Au(001) buffer.

Magneto-optical Kerr spectra in [Fe(nML) /Au(nML)]$_N$ films with thin Fe and Au layers ($1 \leq n \leq 5$) prepared on Au (001) are shown in Fig. 3. The spectral features of these multilayers are completely different from that of n=10. No clear enhancement effect was observed around 2.4 eV. The peak values of rotation below 4 eV do not exhibit a drastic change with layer thickness. A prominent dispersion-type structure centered at 3.6 eV is observed in the Kerr rotation spectrum for n=1, which undergoes a systematic "blue shift" as n is increased. The amount of the energy shift between n=1 and n=5 samples is approximately 1 eV.

Comparison between MO spectra of the sample with n=10 and those with n=1-5 leads to a conclusion that the electronic structures of the Au layer in the Fe/Au multilayers deposited on Au(001) with n=1-5 are not similar to those of thick Au layers which may be assumed to hold the bulk nature.

In order to clarify whether or not the magneto-optical structures observed around 4 eV in Fig. 3 are related to the plasma frequecies, the reflectivity spectra were measured between 0.5 and 7 eV. Some examples of experimental results are illustrated in Fig. 4. The reflectivity spectrum in the Fe(10ML)/ Au(10ML) multilayer shows a distinct break point around 2.4 eV, where the peak of Kerr rotation occurs. On the other hand, the reflectivity undergoes a monotonous decrease up to the highest energy of the measurement in multilayers consisting of thin layers of Fe and Au (n=1 - 5).

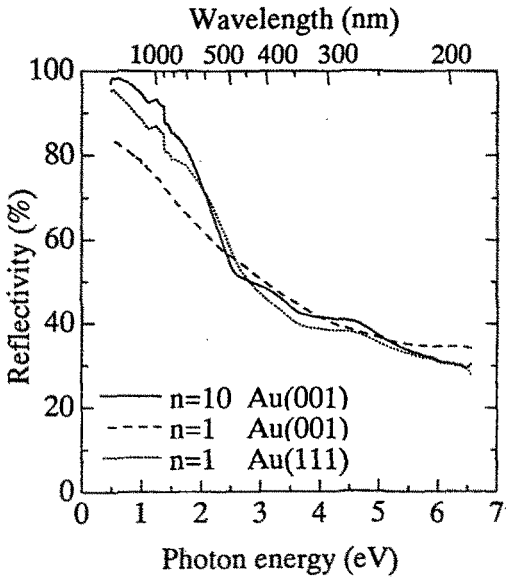

Fig. 4 Typical examples of relectivity spectra of Fe/Au multilayers. Solid curve represents [Fe(10ML)/Au(10ML)]$_{10}$ multilayers on Au(001), dashed curve [Fe(1ML)/Au(1ML)]$_{100}$ on Au (001) and dotted curve [Fe(1ML)/Au(1ML)]$_{100}$ on Au (111).

Real and imaginary parts of the dielectric function were calculated by the Kramers-Kronig analysis of the reflectivity. We used the optical constants (for photon energies between 1.5 and 3

eV) measured by means of spectroscopic ellipsometry to calibrate the values of the dielectric constants.

As a typical example, real and imaginary parts of the diagonal component of dielectric function in [Fe(1ML) /Au(1ML)]$_{100}$ are shown in Fig. 5. No drastic change of spectral features were observed in $n \geq 2$ multilayers. As shown in Fig. 5 real part of diagonal dielectric function ε_{xx} never crosses the abscissa near the photon energy (3.6 eV) corresponding to the peak of Kerr rotation. Thus it is found that the Kerr rotation peak cannot be ascribed to enhancement by plasma resonance.

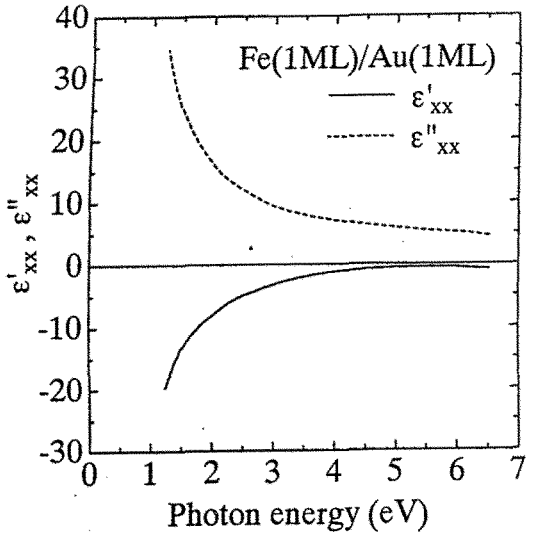

Fig. 5 Spectrum of dielectric function in [Fe(1ML)/Au(1ML)] on Au (001) buffer.

From experimentally obtained data of θ_K, η_K and ε_{xx} we evaluated off-diagonal elements of conductivity multiplied by angular frequency $\omega\sigma_{xy}$, which is known to be useful for discussion of magnetooptical effect in terms of the electronic structures. The spectra are given in Fig. 6.

It should be noted that $\omega\sigma''_{xy}$ takes an approximately constant value ($\sim -2\times10^{29}s^{-2}$) between 1 and 4 eV. Erskine and Stern argued that a constant value of $\omega\sigma''_{xy}$ can be correlated with the spin polarization of conduction electrons. [8] Such behavior has not been observed in the $\omega\sigma''_{xy}$ spectrum of Fe and suggests that electronic states at the Fermi surface in the Fe/Au multilayers are completely different from those of Fe.

On the constant value, a peak of the off-diagonal conductivity is superposed around 4-5eV. We believe that the 4 eV transition originate from band-to-band transition in the new energy band scheme of the superlattice.

To confirm this point we made a rough estimate of $\omega\sigma''_{xy}$ using the density of states (DOS) curve obtained by the electronic band structure calculations in the Fe[1ML] /Au[1ML] superlattice.[9]

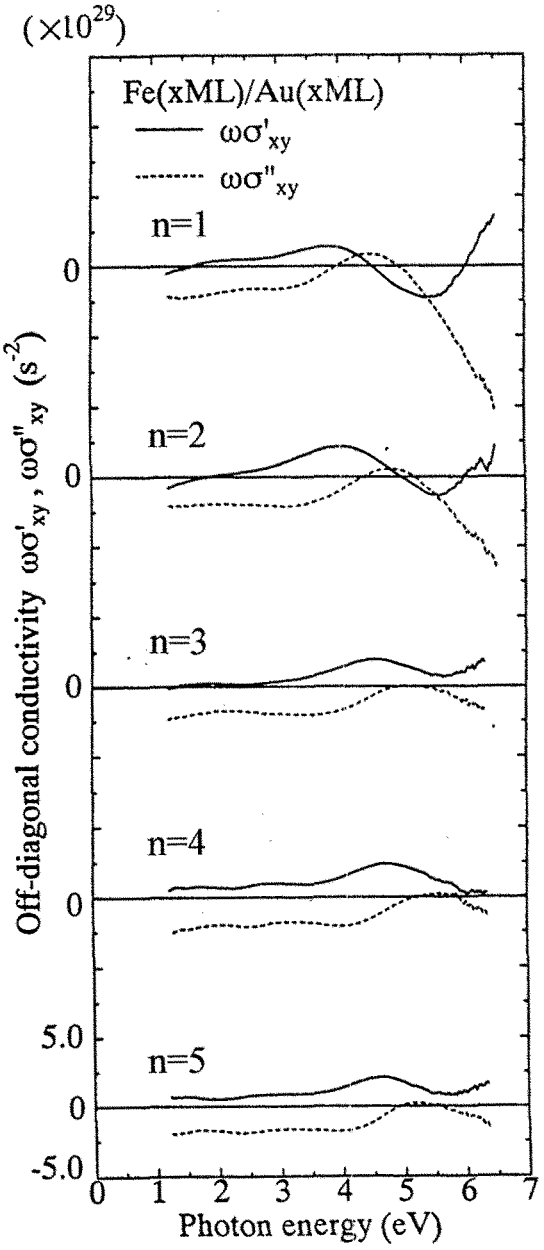

Fig. 6 Spectra of $\omega\sigma''_{xy}$ in [Fe(nML)/Au(nML)]$_N$ multilayers on Au (001).

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The spectrum of $\omega\sigma''_{xy}$ was evaluated by taking an energy-derivative of the joint density of states derived from a convolution between the filled and the empty states in the DOS curves, the same procedure having been described elsewhere.[10] The calculated spectrum of $\omega\sigma''_{xy}$ is shown in Fig. 7. A distinct peak is found around 4 eV in the spectrum, which arises mainly from the filled 5d states (down spin) of Au to the empty 3d states (down spin) of Fe. This result supports the idea that the Fe[1ML]/Au[1ML] superlattice is not a simple stack of Fe and Au layers but a novel ordered alloy between them.

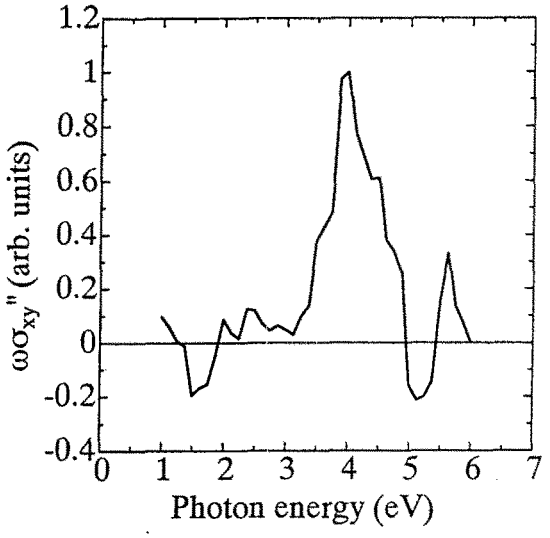

Fig. 7 A spectrum of $\omega\sigma''_{xy}$ estimated from the density of state curve.

We simulated spectra for $n\geq 2$ by the virtual optical constant method assuming that the superlattice consists of interfacial n=1 alloy and "bulk" layers of Fe and Au. The calculated spectrum showed a peak at the plasma edge of Au, while the characteristic dip remained at the same position although the absolute value of rotation is smaller.

Fe/Au MULTILAYERS ON Au (111)

Figures 8(a) and 8(b) illustrates magneto-optical Kerr rotation and Kerr ellipticity in [Fe(nML) /Au(nML)]$_N$ multilayer with n=1, 2 and 3 on Au(111). The absolute values of peak Kerr rotation of these multilayers on Au (111) are approximately twice as large as that of multilayers deposited on Au (001) buffer.

It should also be noted that the multilayer with n=1 shows different MO spectra from the rest. The spectral feature of the former is quite similar to that of n=10 multilayer on Au(001), with a broad peak of Kerr rotation around 2.5 eV, although the absolute value of rotation is smaller.

On the other hand, spectra of [Fe(nML) /Au(nML)]$_N$ multilayer on Au(111) for n=2 and 3 resemble with each other. They both show striking similarity with the MO spectrum of Fe/Au with n=3 on Au(001). This suggests the electronic structures of periodic multilayers on Au (111) are not so much different from those on Au (001).

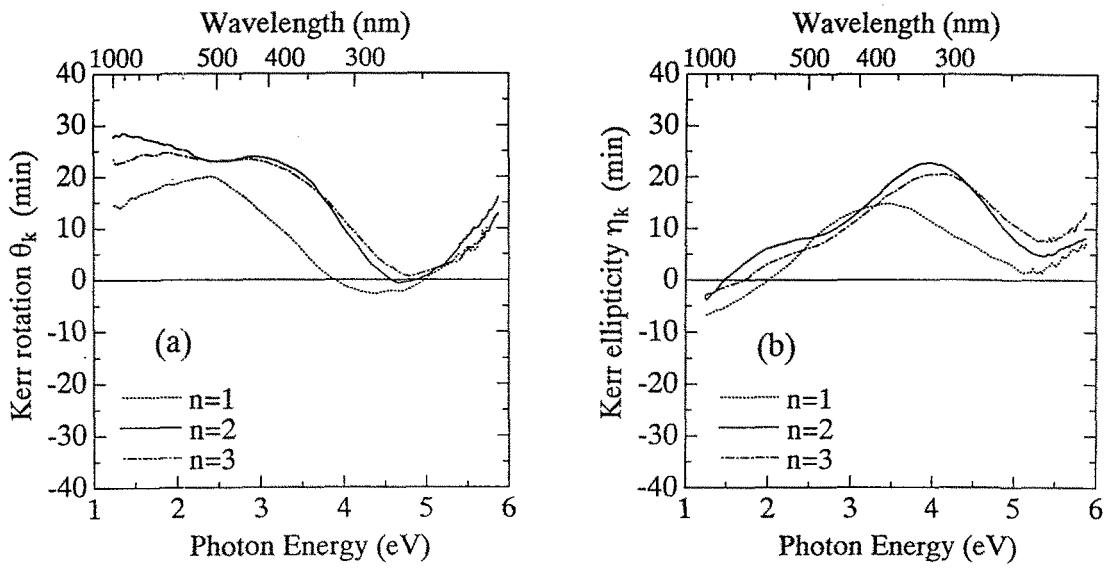

Fig. 8 Spectra of magneto-optical Kerr rotation (a)and Kerr ellipticity (b) in [Fe(nML)/Au(nML)]$_N$ on Au(111) for n=1(dotted), 2 (solid) and 3 (dot-dashed).

As was shown in Fig. 4, the reflectivity spectrum of the n=1 multilayer on Au(111) is quite similar to that of the n=10 film on Au(001), but is different from the n=1 multilayer on Au(001).

As described earlier, the n=1 multilayer on Au(111) does not show any superlattice line in XRD pattern, whereas multilayers with n=2, 3 show a periodic structure. Therefore it may be postulated from these experiments that the electronic structures of multilayers are different from those of a simple stack of two constituent layers as far as an artificial ordered structure is formed.

Lack of periodic structure in the n=1 multilayer may be attributed to inferior surface flatness of the Au(111) surface, for which an improvement of the growth technique should be saught in future studies.

CONCLUSION

From the optical and magneto-optical spectra it has been elucidated that the electronic structures of the [Fe(nML) /Au(nML)]$_N$ multilayer with n=1-5 are completely different from that of a simple stack of Fe and Au layers. The existence of spin-polarized conduction electrons at the Fermi surface is suggested from the spectra of off-diagonal conductivity. Magneto-optical structure around 4eV was attributed to an optical transition between Au 5d and Fe 3d states.

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