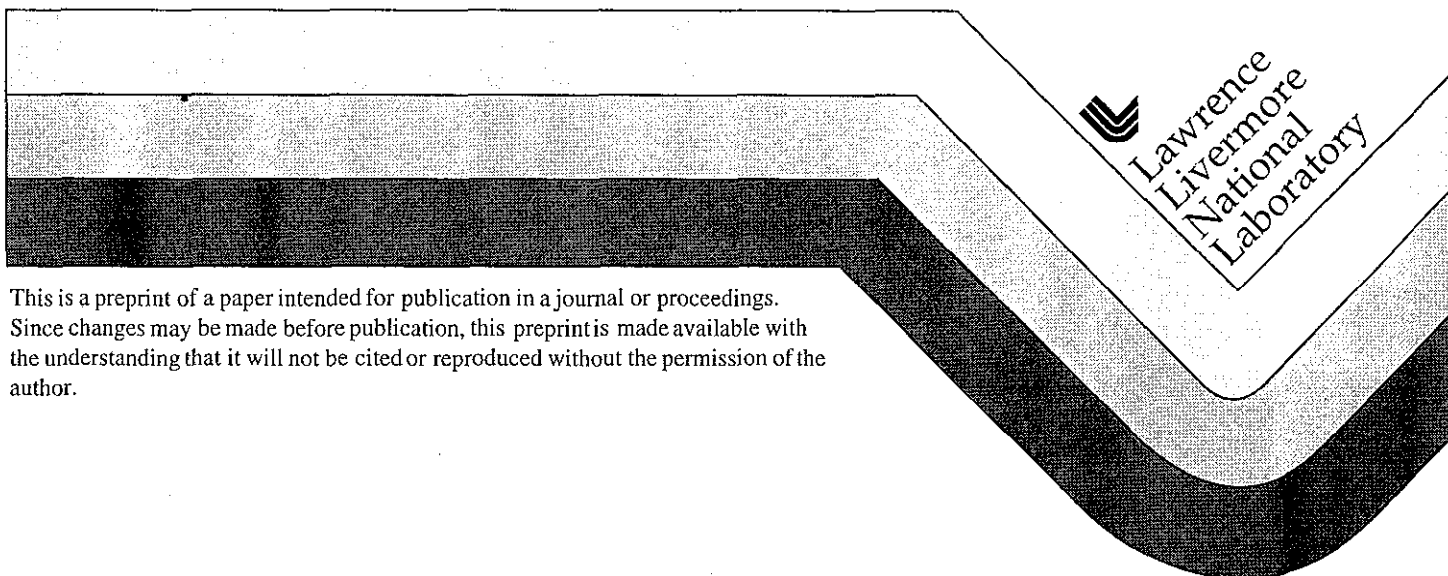# Element-Specific Magnetometry with Linear Dichroism in Photoemission

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Element-specific magnetometry with linear dichroism in photoemission

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Abstract

In this paper, we investigate the magnetic linear dichroism in the core-level photoemission spectra of the binary alloys Co_xNi_{1-x} and Fe_xNi_{1-x}/Cu(100).These epitaxial films have fcc structures, but very different magnetic behavior.We show that the magnetic linear dichroism in x-ray photoemission (XMLD) signal tracks the magnetization in these alloys. Comaprison with recent SQUID data provides a quantitative check and endorses the view that XMLD monitors the element-specific magnetometry.

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Magnetic dichroism in absorption and core-level photoemission has emerged as new tool for probing magnetic properties. Core-level spectroscopy is inherently element-specific and offers the possibility to investigate element-specific magnetic properties. Specifically it has been shown that changes in the magnetization can be tracked element-specifically [1, 2].

However, a lack of consensus on what constitutes the spectroscopic lineshapes questions the applicability of this method. In this letter, we present results which show conclusively that the dichroism of the integrated sums of the spectral lineshape track the changing magnetization in these alloys. Specifically, we report on the concentration dependence of the magnetic dichroism for fcc Co_xNi_{1-x} and Fe_xNi_{1-x} ultrathin alloy films of the 3p core levels in photoemission with linearly polarized light (XMLD). In the case of Co_xNi_{1-x} alloys we have compared the dichroism obtained with circular and linearly polarized light, and observed essentially the same trends in behavior using both methods[3].

We have choosen fcc Co_xNi_{1-x} and Fe_xNi_{1-x} binary alloys because of their very different behavior in the bulk. Co_xNi_{1-x} is structurally and magnetically well-behaved in particular the magnetic moment varies linearly as a function of concentration[4]. This is in sharp contrast to fcc Fe_xNi_{1-x} which displays a magnetic instability at ~65% Fe content[5]. An extended regime of fcc stabilility is possible via epitaxy on Cu(100) [6-8]. If XMLD is indeed a sensitive probe of the element-specific magnetization, we should expect very different behavior for this latter alloy. This has been observed and will be discussed in the following.

Growth and structural aspects of Co_xNi_{1-x} and $Fe_xNi_{1-x}/Cu(100)$ have been thoroughly studied and we found for both systems good epitaxial growth in the fcc structure[7, 8]. The dichroism experiments were performed at the SpectroMicroscopy Facility on Beamline 7 at the Advanced Light Source, Berkeley[9]. For photoemission of the 3p core levels we utilized 190 eV photons (p-polarized) and collected electrons in normal emission with an angular resolution of 2°. The angle of incidence of the photon beam was 60° with respect to the surface normal and the magnetization was in the 'transverse' geometry[8]. A field pulse from a coil near the sample magnetized the sample along the {001} direction.

First we display the concentration dependent 3p magnetic dichroism asymmetry for ~6 ML $Co_xNi_{1-x}/Cu(100)$ in fig.1 [10]. For the Co 3p level we find a constant asymmetry of ~10% which agrees with previous work by Kuch et al.[11], who investigated Co/Cu(100). For the Ni 3p level we observe a small value of ~2%. The results of the XMLD measurement suggest that the magnetic states of the Co and Ni atoms are not changing, since their asymmetry remains constant as the stochiometry is changed. From the bulk we also know that the moments are concentration independent[4]. This might be regarded as evidence that the XMLD asymmetry is tracking a quantity closely related to the element-specific magnetic moment[2].

In fig.2 we show the magnetic dichroism asymmetry for the Fe and Ni 3p levels. We notice a high asymmetry for the Fe 3p level at low Fe concentrations which is strongly reduced at high Fe concentrations. As explained previously [8]we can identify three regimes (I-III) on the basis of the variation of the atomic volume and the Fe magnetic asymmetry[8].

Clearly the concentration dependence is now distinctively different in the Fe_xNi_{1-x} alloy.

We want to discuss now to what extent XMLD can be used for elemental magnetometry of alloys. We were encouraged to pursue this aspect of our work by the results of Sirotti et al. on Fe bulk samples[2]. They compared the low-temperature dependence of the Fe 3p linear dichroism and asymmetry in the the spin-polarization of secondary electrons. The latter is now very well established to be proportional to the magnetization[12]. Sirotti et al. observed that both techniques gave identical results and they concluded that the XMLD asymmetry is indeed proportional to the overall magnetization.

As shown in fig.1, the element-specific magnetic properties of Co and Ni in Co_xNi_{1-x} remain constant. From these data, we find that the average dichroism asymmetry for Co and Ni is 9.8% and 2%, respectively.

We can replot the data of fig.1 by 'calibrating' the average asymmetries in the following way: (i) 9.8 % asymmetry equals 1.7 μ_B for Co and (ii) 2 % asymmetry equals 0.6 μ_B for Ni.

These are the known magnetic moments for bulk Co and Ni[4, 13]. Now we are able to calculate the average moment as a function of concentration for each data point. First we convert the dichroism asymmetry for Co and Ni into an element-specific moment. Secondly we calculate the average moment using the following stochiometric equation:

$$\mu = x \cdot \mu_{Co/Fe}(x) + (1-x) \cdot \mu_{Ni}(x) \tag{1}$$

If we replace the actual data points with the averaged dichroism asymmetry for Co and Ni, the solid line results, plotted in fig. 3. The error bars reflect the uncertainties of the original data in fig.1.

We can repeat this procedure for Fe_xNi_{1-x} alloys, for which we use the following " calibration": (i) 8.5 % asymmetry equals 2.5 μ_B for Fe and (ii) 2% asymmetry equals 0.6 μ_B for Ni. In fig.2 we have shown that the phase diagram of Fe_xNi_{1-x} alloys can be divided into 3 regimes. We concluded that Fe is in a high-spin (HS) state for concentrations up to ~65%. Following the work of Abrikosov et al. we associate this state with an atomic magnetic moment of 2.5 μ_B; for Ni we have used again the bulk value[14]. Again we can calculate the average moment using equation (1). The solid line in fig.4 is the result if we replace the actual data points by the fitted curve in fig.2.

Clearly in order to endorse XMLD as an element-specific magnetometer we need a comparison to results using absolute magnetometer. Recently Freeland et al. provided results to this effect on ultrathin Fe_xNi_{1-x} alloys[15]. They investigated 4 ML thick Fe_xNi_{1-x} multilayers grown mainly on a Cu(111) substrate. Using SQUID magnetometry they have determined absolute values of the average moment. We show their results together with our reploted data from figs. 3 and 4, in fig.5. We have also included also a data point for 6 ML Fe/Cu(100) from a previous publication of the same group[16]. This clearly shows an almost linear increase for Fe concentrations up to ~65%. Extraplotes to a value of ~3 mb in its fcc Fe is in a HS state. Further evidence that the Fe moment stays constant in a HS state up to 65% Fe has been provided by Mössbauer spectroscopy [15].

This work confirms our conclusions based on the high value of the Fe asymmetry up to ~65% Fe content as discussed previously[8].

Going beyond 65% Fe content Freeland et al. observe a strong reduction of the magnetic moment towards smaller values. For pure Fe they find a magnetic moment of ~1 μ_B. In this Fe-rich regime, their data show a considerable scatter of values, presumably due to different amounts of strain.This is particularly true for the (100) oriented thin films. These show a consistently smaller value of the moment than those in the (111) orientation.

The apparent discrepancy between the experimental data points and our work (full line) in fig.5 can be explained as follows. The difference between the 2 data points from samples with the (100) orientation is due to the thickness. Increasing the thickness obviously reduces the average moment due to the relief of volumetric strain, which is consistent with the Mössbauer experiments on Fe/Cu(100) [17]. Thickness dependent magnetic and structural properties are known to exist in the thickness regime 4-7 ML [17-20]. This is also true for Fe_xNi_{1-x}/Cu(100) alloy films as previously reported [8]. We suspect that the Fe_xNi_{1-x}(111) films investigated by Freeland et al. have not the same atomic volume as our films for Fe concentrations larger than 65%. Either the films in the (111) direction have not fully relaxed or there is real difference in the volume instability. As it turns out there is evidence for the latter. The structure of Fe/Cu(111) has been carefully determined by means of LEED I-V [21]. In accord with earlier reports [22] it is found that for thicknesses up to 5 ML a fcc phase exists and we derive an atomic volume of 11.68 $Å^3$ for 5 ML. This value is almost exactly in between 11.4 and 12.1 $Å^3$ which are representative for 2 different magnetic phases in the case of Fe/Cu(100) [20]. We have essentially derived the same values via an extrapolation [8]. Following the work of Keavney et

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al. we associate an atomic volume of 11.4 Å^3 with an average Fe moment of 0.3 μ_B. On the other hand an average moment of ~2 μ_B can has been observed for the HS state[16, 17]. Making the reasonable assumption that the moment is a linear function of the atomic volume[16] we determine an average moment of ~1 μB for fcc Fe/Cu(111). This is in good agreement with the results of Freeland et al.[15].

It is also important to note that Tian et al. do not observe extra spots in the fcc phase contrary to the observation for fcc Fe/Cu(100) [18, 20, 23]. Obviously the structural instabilities and therefore magnetic states do not manifest themselves as clearly for pure Fe/Cu(111) as compared to Fe/Cu(100). Our work and the results of Freeland et al. agree that for up to ~65% Fe content Fe is in a HS state. Beyond this concentration the behavior of the atomic volume for Fe_xNi_{1-x} in the (100) and (111) orientation is different.

So far we have discussed our results in terms of ground state properties. However our measurements have been performed at temperatures in the range 250-300 K. We will show that any effect of temperature is rather minimal and not significant in the present context. The variation of the magnetization for bulk Ni is given in reduced temperature units T/T_c in Kittel[13]. We see for example that for $T/T_c=0.7$ the magnetization is still 80 % of M(0 K). For example we investigated ~6 ML thick Ni/Cu(100) with XMLD and we know that T_c~400 K [24]. This means that 300 K is equal to 0.75 in reduced units. Consequently the error associated with the measurement at 'high' temperatures is much smaller than the error bar for the Ni XMLD asymmetry, see fig.4. It is the latter which results in the rather large error bar of our average moment plot in figs. 3 and 4. Alloying Ni

with Co pushes the $T_c(d)$ dependence quickly up[7]. We find for 6 ML $Co_{10}Ni_{90}$ that 300 K is equivalent to ~0.65 in reduced units. Now the magnetization has only decreased by ~10%. This trend of a decreasing deviation of the magnetization continues if the Co content is increased. Therefore we conclude that the error due to thermal exciations can be neglected and is at all times smaller than the error bar of the average moment, see fig.3. We can now apply the same reasoning for Fe_xNi_{1-x} alloy films. For concentrations up to ~65 % Fe the thermal excitations account for only ~10% decrease in M(0 K).

For concentrations beyond 65% Fe we have to assume a larger deviation. As shown in a previous publication[7] $Fe_{75}Ni_{25}$/Cu(100) alloys show only weak thickness dependence of T_c in the thickness interval ~4-10 ML. The value is around 300 K which is not much higher than 250 K which was the temperature during thickness dependent measurements [8]. We now estimate that the magnetization has decreased by 40 %. Therefore we should replace the value of 0.3 μ_B for pure Fe with 0.5 μ_B. This is still significantly too small when compared with the data of Freeland et al for the (111) orientation. Therefore the remaining differences are mainly due to the different volume instability for Fe_xNi_{1-x}/Cu(111). ·

In summary, we have systematically investigated element-specific magnetic properties with XMLD for two different binary alloys. These results endorse the use of XMLD in photoemission as an element-specific technique for studying ferromagnetism in metastable, binary-alloy, epitaxial films. The difference in the integrated spectral lineshapes when the direction of the magnetization is switched reflects the magnitude of the magnetization. This is clearly reflected in the contrasting behavior of the Co_xNi_{1-x} and

Fe_xNi_{1-x} alloys with changing stochiometry. The observed behavior of the ferromagnetic response suggests a simple summing of the magnetic moments, depending on the alloy composition. This in turn, further underlines the magnetometry deduced from the XMLD signals.

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Figure caption

Fig.1: Concentration dependence of the Co (points) and Ni (squares) 3p asymmetry for 6 ML thick films at 300 K.

Fig.2: Concentration dependence of the Fe 3p and Ni asymmetry at ~300 K, the film thickness was 5.5 -9.0 ML. Solid line is a fit as explained in [8].

Fig.3: Average moment of Co_xNi_{1-x}/Cu(100) alloy films using the 'calibration' as explained in the text.

Fig.4: Average moment of Fe_xNi_{1-x}/Cu(100) alloy films using the 'calibration' as explained in the text.

Fig.5: Comparison of the calibration curve of fig. 4 and the results of Freeland et al.(points/open squares are for (111)/(100) orientation) [15]. Inluded is also a data point from Keavney et al. (square)[16]. Triangle follows from Kümmerle et al. on Fe/Cu(111) [22].
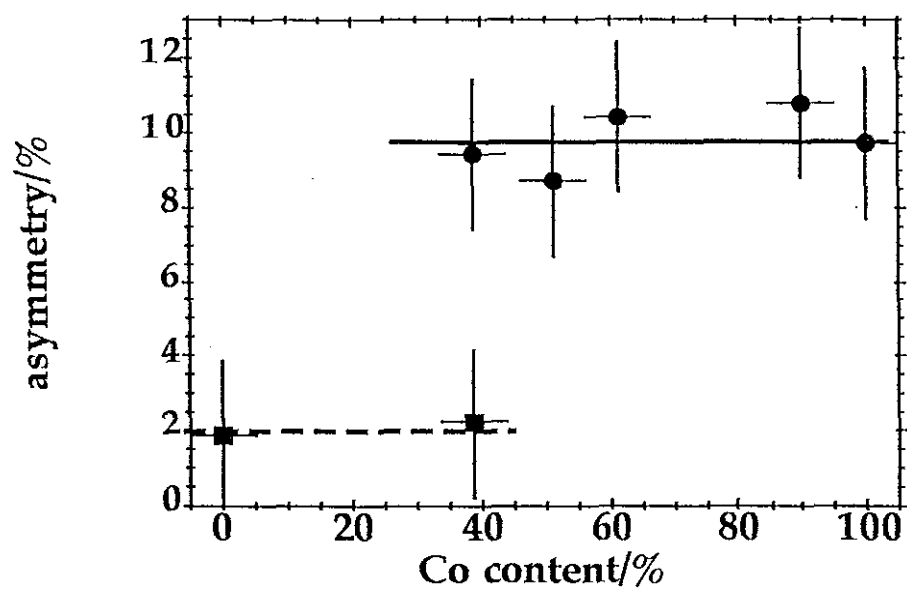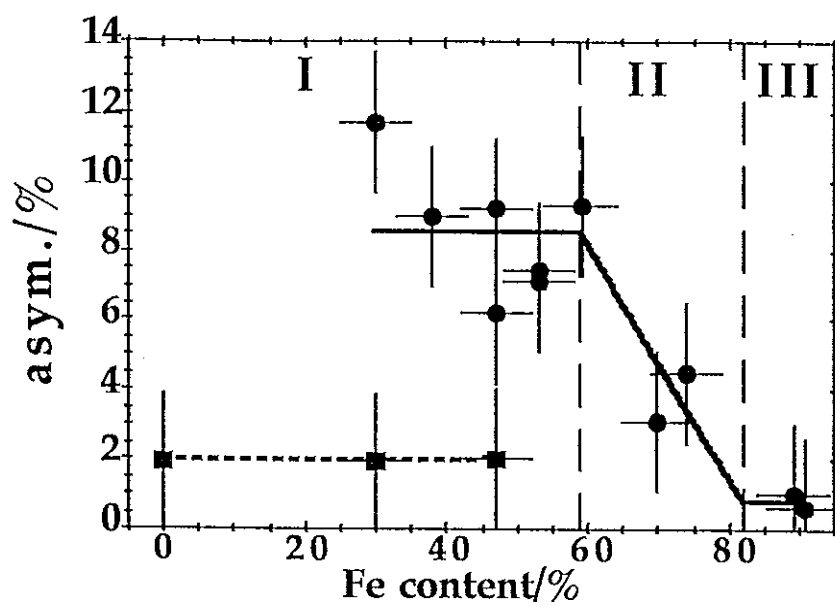
Fig.1

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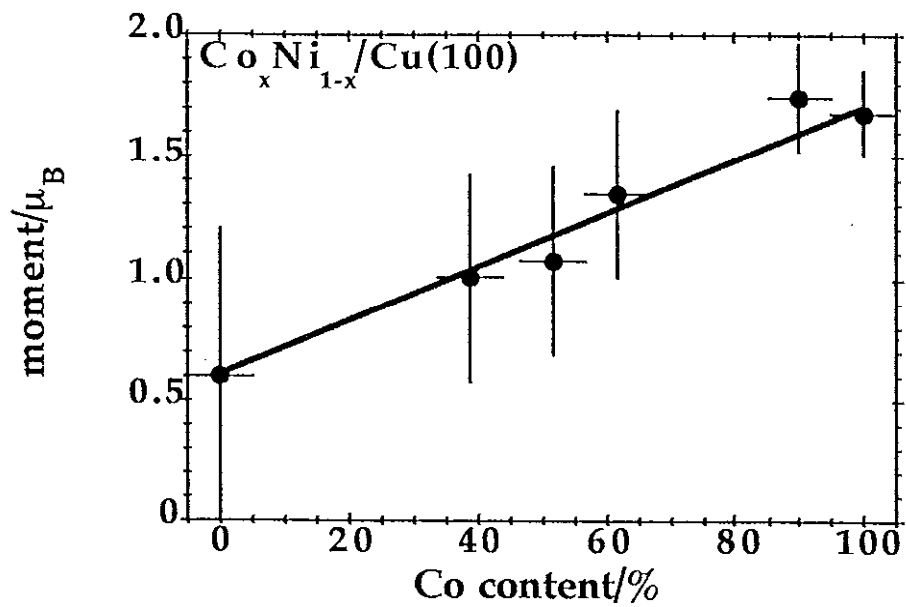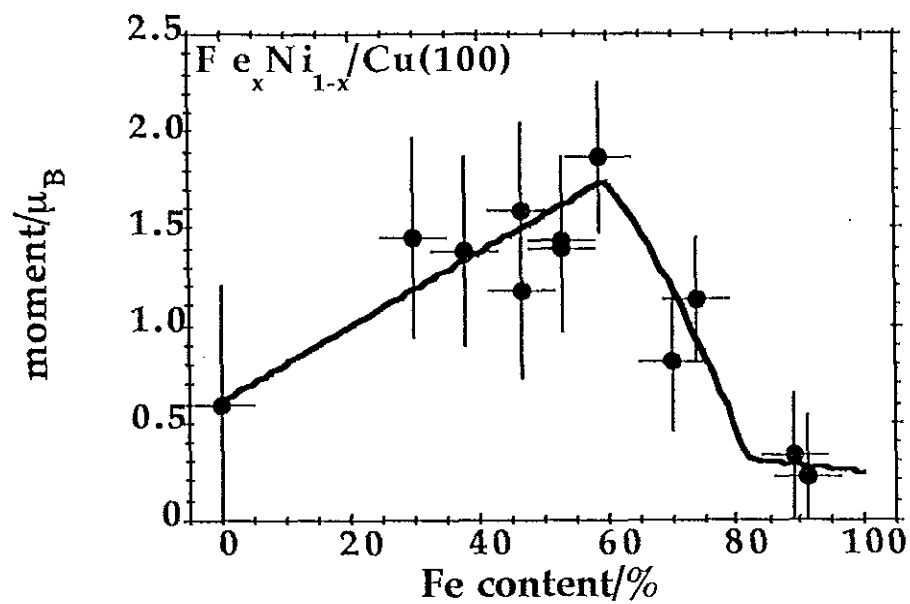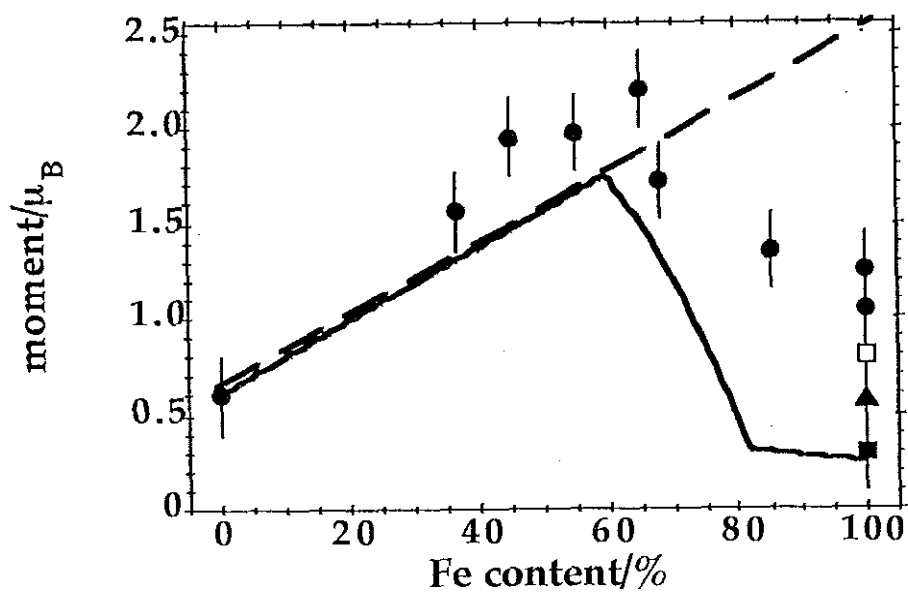
Fig.2

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Fig.5

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