PHYSICAL REVIEW B **75**, 012401 (2007)

Observation of periodic oscillations in magnetization-induced second harmonic generation at the Mn/Co interface

V. K. Valev,[1,2,*] A. Kirilyuk,[1] F. Dalla Longa,[3] J. T. Kohlhepp,[3] B. Koopmans,[3] and Th. Rasing[1]
[1]*Institute for Molecules and Materials, Radboud University Nijmegen, Toernooiveld 1, 6525ED, Nijmegen, The Netherlands*
[2]*Institute for Nanoscale Physics and Chemistry (INPAC), Katholieke Universiteit Leuven, Celestijnenlaan 200D, B-3001 Leuven, Belgium*
[3]*Department of Applied Physics, Center for NanoMaterials (cNM) and COBRA Research Institute, Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands*
(Received 18 October 2006; published 8 January 2007)

Magnetization-induced optical second harmonic generation (MSHG) from exchange-biased Mn/Co thin films shows monolayer period oscillations at the Mn/Co interface as a function of Co thickness. Similar oscillations are found in the exchange bias (H_E) and the coercivity (H_C) in both the interface sensitive MSHG and the bulk sensitive magneto-optical Kerr effect, indicating that magnetic reversal in the Co bulk and at the Mn/Co interface is collinear. Assuming a linear relationship between the MSHG asymmetry and the magnetic moment, our results suggest that there is an enhancement of the interface net magnetic moment at the full monolayer regions.

DOI: 10.1103/PhysRevB.75.012401 PACS number(s): 75.70.Cn, 42.65.Ky

With decreasing material thicknesses, the structural properties of surfaces and interfaces play an increasingly important role in magnetism. Step sites, island sizes, or roughness can have a strong influence upon the magnetic moment, the magnetization reversal behavior, or the magnetic anisotropies, including exchange bias.[1,2]

The latter is obtained when an antiferromagnet (AFM)/ferromagnet (FM) bilayer is cooled from above the Néel temperature of the AFM but below the Curie temperature of the FM, in the presence of an external magnetic field. This phenomenon is a striking example of how dramatically the magnetic characteristics of a bilayer can be influenced by the properties at the AFM/FM interface, however, its precise origin remains unclear.

The main reason for this lack of understanding appears to be the lack of experimental data relevant to the buried AFM/FM interface. Indeed, only a few techniques allow the study of buried magnetic interfaces such as neutron diffraction,[3] magnetic dichroism[4] (or in conjunction with photoemission electron microscopy[5]), and conversion electron Mössbauer spectroscopy,[6] and the studies of exchange bias interfaces using these techniques are often challenging.[7]

Recently, a theoretical model featuring an incomplete domain wall (IDW) developing in the ferromagnet was proposed,[8–10] and it has received significant attention from the scientific community.[11,12] It is characterized by a compensated AFM interface and a canted AFM spin configuration with respect to the direction of the FM spins. The presence of a FM-IDW would affect differently the magnetization reversal process at the AFM/FM interface and in the bulk FM. It has been previously found that magnetization reversal at the surface of a Co/Cu(001) system may differ from the bulk one.[13] It is therefore of great interest to know whether a similar behavior occurs at an exchange biased AFM/Co interface, as that would confirm the validity of the FM-IDW model.

In this paper we have applied the interface-sensitive technique of magnetization-induced second harmonic generation (MSHG) in combination with the magneto-optical Kerr effect (MOKE) to the study of Mn/Co bilayers, where the Co was a layer-by-layer grown wedge on a Cu(001) single crystal and the Mn was deposited as a thin film. Each new Co layer starts by forming islands that (almost) completely fill the surface before the next monolayer starts growing. Thus, the wedge passes through alternating phases of being atomically flat (filled layer) and rough (half filled layer) as can be observed with scanning tunneling microscopy.[14] This system allows us to explore the variation of atomic scale roughness at the AFM/FM interface in a well-controlled way, assuming that no interface smoothing of the rough interface regions occurs due to mechanisms such as heavy interdiffusion or annealing.

Our data, benefiting from the interface sensitivity of MSHG, provide unambiguous evidence that the roughness at the interface remains after capping. These results are obtained from the direct observation of the MSHG intensity produced by dominantly nonmagnetic tensor elements. Furthermore, we find an indication of magnetic moment oscillations at the exchange biased interface between the layer-by-layer grown Co on Cu(001) and the antiferromagnetic Mn. Similar oscillations have been observed previously on clean Co surfaces,[15,16] however to our knowledge this is the first time that their kind is revealed at a *buried* interface. After showing how information on the magnetic moment can be extracted from the oscillations of the MSHG signal, we conclude that the former is maximal at filled monolayers. The origin of these oscillations is then discussed in terms of three possible mechanisms. We estimate that the most likely explanation is that involving a canting of AFM spins that are oriented perpendicular to the Co magnetic moment, in agreement with previous studies.[14] Therefore, since the spin structure of the Mn is that of a compensated AFM/FM interface, our system is ideal for testing the FM-IDW hypothesis.

We report that we find no difference in magnetization reversal between the interface and the Co bulk. Both the interface-sensitive MSHG and the bulk-sensitive MOKE hysteresis loops exhibit the same loop shift and coercivity dependence as function of Co growth. Furthermore, the two techniques present a similar hysteresis loop shape.

The Co(001)/Mn(001) bilayers were epitaxially grown on

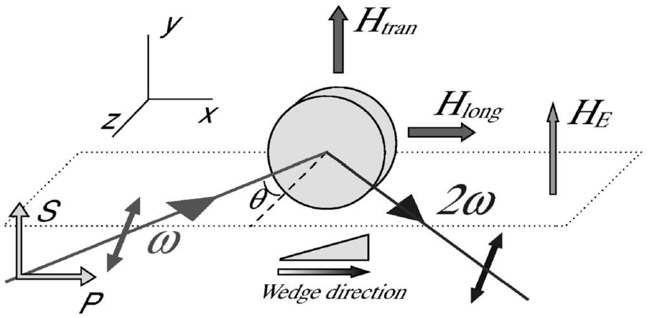

FIG. 1. Experimental configuration. H_{tran} and H_{long} represent the directions of applied field for the transverse and the longitudinal configurations respectively. H_E indicates the direction of the unidirectional anisotropy. The sample was oriented along the Cartesian directions.

atomically clean and flat Cu(001) singe crystals (miscut $<0.1°$) at 330 K in a multichamber molecular beam epitaxy (MBE) system (VG-Semicon V80M) with a base pressure better than 1×10^{-11} mbar. The Co layers were deposited in a wedge structure (roughly 9–13 ML thick) using an *e*-gun evaporator with feedback control of the flux whereas the 25 ML thick Mn films were prepared using a temperature stabilized and extensively degassed Knudsen cell. All nominal thicknesses were controlled by calibrated quartz-crystal monitors, with an accuracy of roughly 3%. During the growth, the pressure never rose above 5×10^{-11} mbar and the growth rates were 1–2 monolayers (ML)/min. Under these conditions, it was shown that the Mn adopts face-centered-tetragonal (fct) structure with a c/a ratio of roughly 1.05, before it transforms around 50–60 ML to the thermodynamically stable complex α-Mn structure.[17] The fct-Mn(001) is antiferromagnetic even at room temperature and is able to induce a sizable exchange anisotropy in the Co layers.[18] To avoid oxidation of the layer during the ex-situ experiments, the bilayers were covered with a 5 nm thick Cu capping layer.

MSHG measurements were performed using a Ti:Sapphire laser at 800 nm wavelength with a pulse width of ~100 fs and a repetition rate of 82 MHz. The laser power was between 20 and 40 mW and the light was focused to a spot with diameter of around 100 μm. The angle of incidence θ was 45° and the magnetic field was applied in the longitudinal and the transverse configurations (see Fig. 1). All the measurements presented in this paper were done at room temperature. The wedge thickness was determined from markers signifying the beginning and end of the wedge.

For intense electromagnetic fields, such as those generated by a pulsed laser beam $\mathbf{E}(\omega)$ incident on a thin multilayer film, the polarization at the harmonic frequency 2ω is given in the electric dipole approximation by

$$\mathbf{P}_i(2\omega)=\chi_{ijk}\mathbf{E}_j(\omega)\mathbf{E}_k(\omega), \tag{1}$$

where χ_{ijk} is a third order polar tensor describing the nonlinear second order optical susceptibility at the symmetry breaking interface between the centrosymmetric films and *i*, *j*, and *k* are the Cartesian indexes.[19]

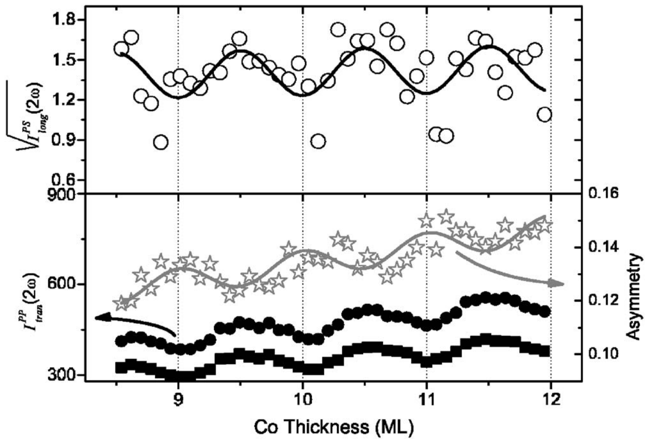

FIG. 2. MSHG intensity as function of Co thickness. Empty circles: the square root of the MSHG intensity for the *P*-*S* polarizer-analyzer combination in the longitudinal magnetic field geometry. The MSHG intensity for the *P*-*P* polarizer-analyzer combination in the transversal magnetic field geometry for positive (full circles) and negative (full squares) magnetic field. The MSHG asymmetry as function of Co thickness is represented by the empty stars.

For the *P*-*P* polarizer-analyzer combination, in the transverse magnetic field geometry, the MSHG intensity is given by $I^{PP}_{tran}(2\omega)\propto|\chi^{even}_{eff}\pm\chi^{odd}_{eff}|^2$, where the superscripts "even" and "odd" indicate that the tensor elements *do not change sign* or *do change sign*, respectively, upon magnetization reversal, and the index "eff" takes into account the Fresnel coefficients and designates the summation over all the tensor elements of identical magnetic characteristic, i.e., all "even" or "odd". *P* represents optical polarization parallel to the plane of incidence. We can then define the average MSHG intensity and asymmetry as

$$I^{PP}(2\omega)=\frac{I^{\uparrow}+I^{\downarrow}}{2}\propto|\chi^{even}_{eff}|^2+|\chi^{odd}_{eff}|^2\approx|\chi^{even}_{eff}|^2, \tag{2}$$

$$A=\frac{I^{\uparrow}-I^{\downarrow}}{I^{\uparrow}+I^{\downarrow}}\propto\frac{|\chi^{odd}_{eff}||\chi^{even}_{eff}|}{|\chi^{odd}_{eff}|^2+|\chi^{even}_{eff}|^2}\cos\varphi\approx\frac{|\chi^{odd}_{eff}|}{|\chi^{even}_{eff}|}\cos\varphi, \tag{3}$$

where φ is the phase difference between χ^{odd}_{eff} and χ^{even}_{eff}, and the quantities $I^{\uparrow}$ and $I^{\downarrow}$ are the MSHG intensities for opposite directions of the magnetization. From Eq. (2), it follows that for $|\chi^{even}_{eff}|\gg|\chi^{odd}_{eff}|$, the MSHG intensity in the *P*-*P* configuration measures the structural properties of the interface.

In Fig. 2, the MSHG intensity is plotted as function of increasing Co thickness. It is clear that the oscillations of I^{PP} present maxima at half filled monolayers, and therefore appear to be related to the interface roughness. This is consistent with a signal that is dominantly originating from the top Co interface since in systems with inversion symmetry the MSHG is generated in regions where this symmetry is broken, i.e., the regions with higher roughness.[20,21] The amplitude of the oscillations is approximately 20%; however one should take into account that there is a contribution to the observed signal by the second Co interface, but of opposite phase. Therefore it is difficult to extract quantitative

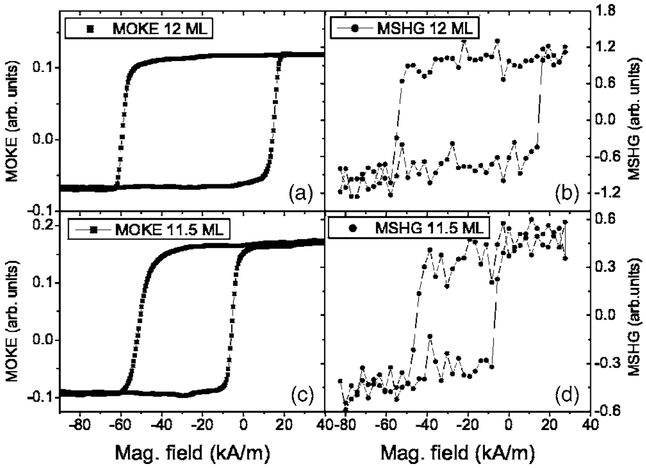

FIG. 3. Hysteresis loops from MOKE (left panels) and MSHG (right panels) taken at thicknesses of 12 ML and 11.5 ML of Co.

information from this amplitude since the detected MSHG is affected by interference between these interfaces. The increase of the total I^{PP} in Fig. 2 results from the fact that due to the increasing Co thickness the signal generated at the second Co interface diminishes, although very slightly.

A further confirmation that the oscillations in I^{PP} should be attributed to the interface roughness comes from examining the magnetization contribution to the odd tensor components. For the *P*-*S* polarizer-analyzer combination, in the longitudinal magnetic field geometry, the MSHG intensity for the effective values of the susceptibilities is given by $I_{long}^{PS}(2\omega) \propto |\chi_{yxx}^{odd} + \chi_{yzz}^{odd}|^2$. Consequently, $\sqrt{I_{long}^{PS}(2\omega)}$ is proportional to χ_{eff}^{odd}. However, note that although χ_{eff}^{odd} is directly proportional to the magnetization,[16] it can and most likely will also be affected by the local electromagnetic fields (LEF) and local electronic structure (LES).[22,23] The later two are different at island edges and thereby can contribute to the oscillations of χ_{eff}^{odd}. Henceforth, we believe that extracting purely magnetic information from the variations of the odd components alone is impossible as both the LEF and the LES on the one hand and the magnetic moment on the other hand could be oscillating.

In Fig. 2, we observe that the monolayer oscillations of $\sqrt{I_{long}^{PS}(2\omega)}$ are in phase with those of the roughness, i.e., maxima occur at half filled monolayers. In order to make sure that the measurement had no "contamination" from even tensor components, the magnetic contrast was measured and found to be zero. From this we can conclude that either the LEF and the LES contributions to χ_{eff}^{odd} dominate or that both these contributions and the magnetic moment oscillate in phase with the roughness.

In order to lift this ambiguity, we examined the magnetic asymmetry. From Eq. (3), it follows that *A* is proportional to the ratio of odd tensor elements divided by the even ones. Assuming that the nonvanishing even and odd components are similarly affected by the LEF and LES, this quantity is to a first approximation only proportional to the magnetic moment [see Eq. (19) in Ref. 22 or Eq. (24) in Ref. 23].

In Fig. 2, we can see that the asymmetry exhibits clear monolayer oscillations but with opposite phase with respect to the interface roughness. Consequently, we can conclude that the interface net magnetic moment is maximal at the flat regions of the interface. Furthermore, since the magnetic moment does not oscillate in phase with the LEF and the LES, to a first approximation, we can attribute the oscillations of χ_{eff}^{odd} to these two quantities.

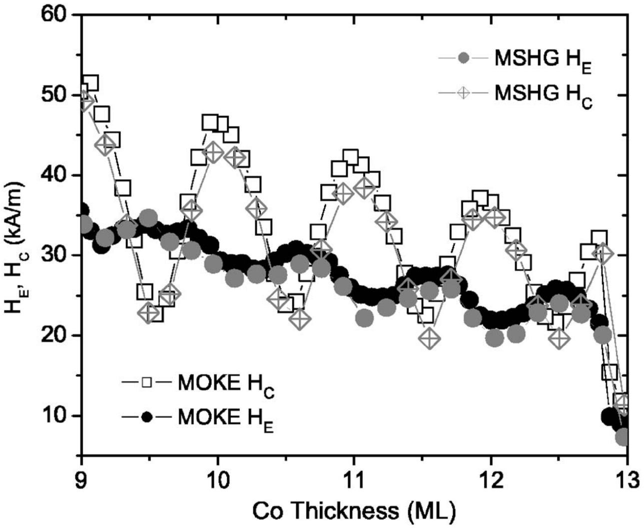

FIG. 4. Coercivity and loop shift of the hysteresis curves from MSHG (in gray) and MOKE (in black) as function of the Co thickness. The last two points signify the end of the wedge.

The oscillations of the interface net magnetic moment could be due to inhomogeneities across the Co thin film. For instance, it is conceivable that under the influence of roughness the magnetization reversal at the interface is incomplete or differs from that of the bulk. One could certainly expect such mechanisms to occur in the context of the FM-IDW model.

To compare the bulk magnetic properties with those of the interface, we measured the MSHG and MOKE hysteresis loops at filled and half-filled monolayers. In Fig. 3 we can see that both hysteresis loops exhibit the same loop shape indicating similar magnetization reversal behavior.

This is further confirmed when we examine the values of the coercivity H_C and the loop shift H_E as function of the Co thickness (see Fig. 4).

Indeed both techniques reveal the same characteristic behavior and the small differences observed are likely due to slightly different calibrations of the MSHG and MOKE setups or to a small temperature increase from the higher laser power necessary for MSHG. Since we have demonstrated that the MSHG signal originates from the interface (the oscillations from crystallographic and magnetic origin exhibit monolayer-periodicity similarly to the interface roughness), we can conclude that the bulk and the interface Co spins behave in the same way, i.e., that there is no difference in magnetization reversal between the bulk and the interface.

These results are of importance for the understanding of exchange bias, since they exclude the possibility of a domain wall formation in the ferromagnet under the influence of the pinning of the antiferromagnet as it is suggested in the FM-IDW model. We believe that, as the Co-Co inner exchange coupling is much larger than the interfacial exchange, if a domain wall is indeed formed, it is more likely to be situated in the antiferromagnet.[24]

Additionally, explaining the enhancement of the net magnetic moment that we observe would be of great interest. However, in the case of an interface this explanation is significantly more difficult than in the case of a surface. Indeed, while it has been shown that there is an increased magnetic moment on the island edges of the Co surface,[15] we believe that this explanation cannot be retained for the interface because of the presence of Mn atoms. On the other hand, variations in the strain/stress conditions between flat and rough regions of the Mn/Co interface should be considered as a possible mechanism.[16] To our knowledge there is no theoretical work that has addressed this problem in the case of a Mn/Co bilayer and, in this particular case, it is clear that the analysis has to include the exchange bias interaction.

A relationship between an enhancement of the interfacial net magnetic moment and the presence of AFM pinned uncompensated spins should not be completely excluded. Indeed, we have demonstrated previously that the presence of such spins can affect strongly the MSHG signal.[25] Nevertheless the experiments that we describe in this paper are not sensitive to *pinned* uncompensated spins, since in the MSHG asymmetry the magnetic moment is a quantity that changes sign upon magnetization reversal. This is confirmed by the fact that the oscillations of the exchange bias and those of the net magnetic moment have opposite phases. Therefore, we believe that this hypothesis is unlikely.

Instead, our experiment could reveal AFM uncompensated (but not *pinned*) spins that are strongly coupled to the FM ones and that reverse with them, thereby contributing to an enhancement of the interfacial net magnetic moment. It has been found that this type of AFM uncompensated spins is responsible for the enlargement of the coercivity in exchange biased systems.[26] We would therefore expect a similar behavior between the coercivity and the net magnetic moment, and this would lead to maxima in the MSHG oscillations at the flat regions of the interface, in accordance with our observations. The simultaneous enhancement in net magnetic moment and coercivity is therefore consistent with the presence of Mn uncompensated spins at the exchange-biased interface. However, we see no reason why more uncompensated spins would form at the flat regions of the interface; in fact intuitively we would expect the contrary.

Consequently, a canting of the AFM spins at the interface is most likely responsible for an enhancement of the interface net magnetic moment. Such a canting of the interface spins associated with an almost orthogonal alignment of the FM and AFM interface spins was actually proposed earlier for this system.[14] Reversing the magnetization is then accompanied by an inversion of the canting angle and thereby this process can contribute to the MSHG asymmetry. This is further supported by the fact that within the biquadratic coupling model, the canting has been suggested to cause an enhancement of coercivity,[27,28] which is again consistent with our observation that the oscillations of the MSHG asymmetry and the coercivity are in phase. As to the micromagnetic reason, it is possible that at certain defect sites at the rough regions of the interface the AFM interfacial order is disturbed. This could prevent the canting at these particular locations, resulting in an average diminishing of the net magnetic moment at half filled monolayers.

In conclusion, a comparison of the interface-sensitive MSHG and the bulk-sensitive MOKE hysteresis loops reveals that there is no difference in magnetization reversal between the Mn/Co interface and the Co bulk, where the Mn spin order is that of a compensated interface, in a canted spin configuration with respect to the direction of the FM spins. This finding is based on an unambiguous evidence that the roughness at the topmost monolayer of Co from a layer-by-layer grown Co/Cu(001) is preserved after capping with Mn, and on a direct observation of the effect of this roughness on the net magnetic moment of the exchange biased Mn/Co interface with MSHG. After careful analysis, within the limits of our assumptions, we can conclude that the interfacial net magnetic moment is maximal at the flat interface regions consistently with a canting of the AFM spins at the interface.

This work was supported in part by the EU projects DYNAMICS and ULTRASWITCH, NanoNed, and the Dutch organization for Fundamental Research on Matter (FOM).

---

*Electronic address: V.Valev@science.ru.nl

[1] W. H. Meiklejohn and C. P. Bean, Phys. Rev. **102**, 1413 (1956).

[2] J. Nogués and I. K. Schuller, J. Magn. Magn. Mater. **192**, 203 (1998).

[3] H. Zabel *et al.*, Physica B **276**, 17 (2000).

[4] J. B. Kortright *et al.*, J. Magn. Magn. Mater. **207**, 7 (1999).

[5] B. P. Tonner *et al.*, J. Electron Spectrosc. Relat. Phenom. **78**, 13 (1996).

[6] T. Shinjo and W. Keune, J. Magn. Magn. Mater. **200**, 598 (1999).

[7] W. A. A. Macedo *et al.*, Phys. Rev. B **70**, 224414 (2004).

[8] M. Kiwi *et al.*, Europhys. Lett. **48**, 573 (1999).

[9] M. Kiwi *et al.*, Appl. Phys. Lett. **75**, 3995 (1999).

[10] J. Meija-Lopez *et al.*, J. Magn. Magn. Mater. **241**, 364 (2002).

[11] Z.-P. Li *et al.*, Phys. Rev. Lett. **96**, 217205 (2006).

[12] S. Roy *et al.*, Phys. Rev. Lett. **95**, 047201 (2005).

[13] M. Gruyters *et al.*, Phys. Rev. Lett. **94**, 227205 (2005).

[14] J. T. Kohlhepp *et al.*, J. Magn. Magn. Mater. **286**, 220 (2005).

[15] Q. Y. Jin *et al.*, Phys. Rev. Lett. **80**, 4056 (1998).

[16] M. Nyvlt *et al.*, Phys. Rev. Lett. **95**, 127201 (2005).

[17] J. T. Kohlhepp and W. J. M. de Jonge, Phys. Rev. Lett. **96**, 237201 (2006).

[18] J. T. Kohlhepp *et al.*, Appl. Phys. Lett. **89**, 032507 (2006).

[19] A. Kirilyuk and Th. Rasing, J. Opt. Soc. Am. B **22**, 148 (2005).

[20] A. Kirilyuk *et al.*, Appl. Phys. Lett. **72**, 2331 (1998).

[21] G. Tessier and P. Beauvillain, Appl. Surf. Sci. **164**, 175 (2000).

[22] S. B. Borisov and I. L. Lyubchanskii, Opt. Spektrosk. **61**, 1274 (1986) [Opt. Spectrosc. **61**, 801 (1986)].

[23] Ru-Pin Pan *et al.*, Phys. Rev. B **39**, 1229 (1989).

[24] U. Schlickum *et al.*, Phys. Rev. Lett. **92**, 107203 (2004).

[25] V. K. Valev *et al.*, Phys. Rev. Lett. **96**, 067206 (2006).

[26] H. Ohldag *et al.*, Phys. Rev. Lett. **87**, 247201 (2001).

[27] N. C. Koon, Phys. Rev. Lett. **78**, 4865 (1997).

[28] T. C. Schulthess and W. H. Butler, Phys. Rev. Lett. **81**, 4516 (1998).