PHYSICAL REVIEW B **68**, 060102(R) (2003)

Non-Gilbert-type damping of the magnetic relaxation in ultrathin ferromagnets: Importance of magnon-magnon scattering

J. Lindner,* K. Lenz, E. Kosubek, and K. Baberschke
Institut für Experimentalphysik, Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany

D. Spoddig, R. Meckenstock, and J. Pelzl
Experimentalphysik III, Ruhr-Universtät Bochum, Universitätsstraße 150, D-44801 Bochum, Germany

Z. Frait
Institute of Physics, Academy of Sciences of Czech Republic, 18040 Prague, Czech Republic

D. L. Mills
Department of Physics and Astronomy, University of California, Irvine, California 92697, USA
(Received 20 June 2003; published 25 August 2003)

Ferromagnetic resonance (FMR) measured over a large range of frequencies from 1–70 GHz offers a unique possibility to study the dynamic response of ultrathin ferromagnetic films in the range from nanoseconds to picoseconds. The linewidth of the FMR signal is commonly believed to follow a linear ω dependence, the so-called Gilbert damping. Here we give experimental unambiguous evidence that other processes of spin dynamics such as two-magnon scattering are equally important at interfaces of ferromagnetic to nonmagnetic nanostructures. The relevance to spin transport and spin injection as well as the agreement with recent theoretical proposals are discussed.

DOI: 10.1103/PhysRevB.68.060102 PACS number(s): 75.70.Cn, 76.50.+g

The physics of magnetism in ultrathin films and in multilayers formed from such ultrathin films has proved fascinating, since one encounters diverse new phenomena not present in bulk magnetic materials. For example, ultrathin films are realizations of two-dimensional magnetic matter, and in fact by varying their thickness, one may follow the transition from two- to three-dimensional magnetism.[1,2] In multilayers, oscillatory exchange coupling between neighboring films, very weak in strength compared to interatomic exchange in bulk materials, allows one to fabricate spin valves whose state is altered in very weak external fields,[3] and exotic magnetic phases in more extended superlattice structures, also induced by modest applied fields.[4] Furthermore in the spin dynamics of ultrathin structures, one encounters novel physics. In particular, new mechanisms of spin damping have been observed which are intimately related to the ultrathin film character of the samples.

Two new mechanisms have been the topic of recent discussion concerning the damping at interfaces of ferromagnetic (F) to nonmagnetic (N) materials:

(i) The two-magnon mechanism, with origin in surface or interface defects.[5]

(ii) The transfer of angular momentum from the precessing magnetization excited, say in ferromagnetic resonance (FMR) experiments, to the conduction electrons in these metallic films.[6]

As a consequence the magnetization acts as a spin pump. Hence, the angular momentum so transferred is transported out of the film, or otherwise dissipated.

In both bulk materials and in ultrathin films, for many decades and throughout the basic and applied literature on magnetization dynamics motions of the magnetization are described through use of the classical Landau-Lifshitz-Gilbert equation, written as[7,8]

$$\frac{1}{\gamma}\frac{\partial \mathbf{M}}{\partial t} = -(\mathbf{M}\times\mathbf{H}_{\mathrm{eff}}) + \frac{G}{\gamma M_s^2}\left(\mathbf{M}\times\frac{\partial \mathbf{M}}{\partial t}\right). \tag{1}$$

Here the first term describes the precession of the magnetization in the effective fields with origin in several sources, including the externally applied Zeeman field, anisotropy fields, and exchange fields in circumstances where the magnetization being excited is spatially non-uniform. In addition, the microwave field of frequency $\omega/2\pi$ applied in FMR is included here. The data reported here show that even for small amplitude spin motions about the equilibrium orientation, this widely used phenomenological form fails to describe the response of the spin system. The second term in Eq. (1) describes damping of the spin motions and leads to the linewidths observed in FMR experiments. This term and its implications are the focus of the present Rapid Communication, and the data we show illustrate clearly that it fails to describe the linewidths we observe in our measurements, which are the first to be presented over a wide range of frequencies. The time-derivative $\partial\mathbf{M}/\partial t$ in the Gilbert term produces a FMR linewidth linear in the resonance frequency ω,

$$\Delta H_{\mathrm{Gilb}}(\omega) = 1.16\frac{\omega G}{\gamma^2 M_s}. \tag{2}$$

The notion that the linewidth scales linearly in frequency is employed very widely in the literature. It is also the case that the ''spin pumping'' mechanisms discussed recently in Ref. 6 also were shown to lead to a linear variation of the linewidth with frequency and thus these effects can be incorporated into an effective Gilbert-damping constant G. However, in ultrathin films, while a linear variation is reported commonly, the linewidth fails to extrapolate to zero with vanishing frequency. This has led to the concept of a ''zero-field linewidth'' ΔH_0 which should be added to Eq. (2).[9] Its origin is assumed to reside in inhomogeneous broadening of the FMR line. The focus of the present Rapid Communication is therefore to question the proportionality of the linewidth to the driving frequency ω. We will give an experimental evidence here that other damping processes in ultrathin films lead to striking and qualitative deviations from the linear frequency dependence provided when the Gilbert-damping term only is operative. The frequency dependence found for the new contribution, as well as a strong fourfold in-plane anisotropy are compatible with the two-magnon mechanism. Our data then raise a most important question: What is the structure of the appropriate phenomenology to describe the magnetization dynamics in ultrathin ferromagnetic films? A possible alternative is the Bloembergen ansatz[10] where separate longitudinal and transverse relaxation times are used to model the leakage of M_z and scattering processes which lead to the decay of $M_{x,y}$. We note in this regard that two-magnon scattering is a dephasing process[5] which leaves M_z unaffected. The issue we raise is important for both the understanding of the basic physics of ultrathin film systems, and for devices that incorporate such films.

The two-magnon mechanism mentioned above predicts that the linewidth does not vary linearly with frequency, but saturates at high frequency.[5] The result in Ref. 5 may be cast into the form

$$\Delta H_{2\mathrm{mag}}(\omega)=\Gamma \sin^{-1}\sqrt{\frac{[\omega^2+(\omega_0/2)^2]^{1/2}-\omega_0/2}{[\omega^2+(\omega_0/2)^2]^{1/2}+\omega_0/2}}, \quad (3)$$

where $\omega_0=\gamma(2K_{2\perp}-4\pi M_s)$, $K_{2\perp}$ being the uniaxial anisotropy constant and M_s the saturation magnetization. One sees that $\Delta H_{2\mathrm{mag}}(\omega)$ vanishes linearly with frequency, but exhibits substantial deviations from linear behavior in the FMR frequency range, for typical parameters. The mechanism is operative because there are short-wavelength magnons degenerate with the uniform mode excited in FMR, regarded as a zero-wave-vector magnon. Defects at interfaces can then scatter energy from the FMR mode to the degenerate short-wavelength modes. For ultrathin films the dipolar energy in a ferromagnet produces a term in the dispersion relation *linear* in the wave vector with negative slope. This is added to the conventional term of exchange interaction with a *positive quadratic* term in the wave vector, and results in the degeneracy just mentioned. Two-magnon scattering has also been discussed before by Patton and co-workers[11] as well as by Heinrich and co-authors.[12] However, most of the previous work, in particular concerning ultrathin magnetic multilayers, was always based on the assumption of a Gilbert-like ω-proportional damping constant.[12]

To distinguish between a linear form $A+B\omega$ fitted to data taken over a limited frequency range and the expression derived in Ref. 5, one requires data over a very wide range of frequencies. In this Rapid Communication we report the measurements of FMR linewidths in ultrathin film samples

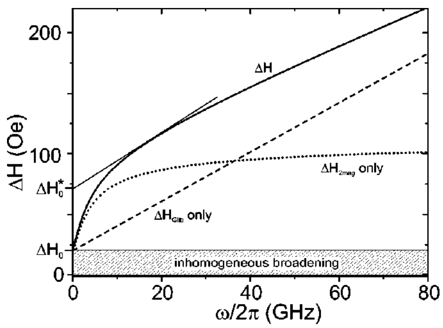

FIG. 1. Schematical diagram for the frequency dependence of a FMR linewidth. The relevant frequency ranges from 1–80 GHz. For the indicated linewidth ΔH ranging from 50 to a few 100 Oe typical FMR parameters were used. Dashed line corresponds to Eq. (2), dotted to Eq. (3), and thick solid line to Eq. (4). The thin solid line indicates one possible tangent leading to an apparent zero-frequency linewidth ΔH_0^*.

using a frequency range of 1–70 GHz, and we find that the data are accounted for very well by combining intrinsic damping of the Gilbert form with the prediction of two-magnon theory. We illustrate the issues schematically in Fig. 1. The dashed line shows the Gilbert damping and its linear frequency dependence. In earlier discussions, the slope was determined from measurements at only two or three frequencies, typically in the 10–36 GHz range. The dotted line is a simulation from Eq. (3) only. In the solid line, we combine the relaxation processes. Clearly, measurements at low frequency, 1–9 GHz, are required to determine the low-frequency behavior of the linewidth. Assembling the entire picture also requires measurements at high frequencies, above 50 GHz. Of course there may be also inhomogeneous broadening present (hatched area), which will manifest itself as a zero field linewidth ΔH_0. There is no spatial resolution in FMR, and the length scales relevant to the measurement are macroscopic, so inhomogeneities in magnetization or anisotropy fields will appear as an apparent frequency independent contribution to the linewidth. Note in Fig. 1 that measurements performed over a limited frequency range could be fitted by a linear function. We illustrate this by the thin solid line, tangent to the full curve. Extrapolation of this to zero frequency produces a fictitious ''zero field linewidth'' ΔH_0^* very much larger than its actual value. It is the case that inferences on the physics of the linewidth in such circumstances can lead to erroneous conclusions, with an incorrect overestimate of the relaxation.

To give evidence for more complicated magnetization dynamics and relaxation mechanisms at *F*-*N* interfaces in nanostructures we have chosen $Fe_n/V_m(001)$ superlattices (SL's) which can be pseudomorphically grown on MgO(001) leading to a high structural as well as magnetic homogeneity. Various groups have used, prepared, and analyzed these Fe/V-SL's on MgO.[13–16] Magnetization and FMR measurements have been also performed so that *g* value, saturation magnetization, and anisotropy constants up to fourth order are well determined in this system.[14,17] The present experiments were performed partly at FUB and the Czech Academy of Sci-

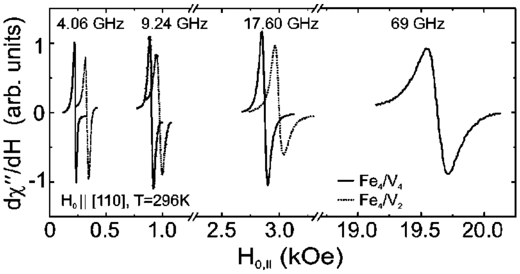

FIG. 2. Typical FMR spectra measured at four different frequencies at room temperature for $(Fe_4/V_4)_{45}$ (solid lines) and $(Fe_4/V_2)_{60}$ (dotted lines). The external field $\mathbf{H}_0$ was oriented along the $[110]$ direction being the hard in-plane axis.

ences at a fixed temperature of $T=296$ K using different FMR setups with frequencies from 1–70 GHz. Two particular samples $(Fe_4/V_2)_{60}$ and $(Fe_4/V_4)_{45}$ were chosen having 60 repetitions of 4 ML (ML, monolayers) Fe and 2 ML V and 45 repetitions with 4 ML V, respectively. For both samples the easy axis of the magnetization lies in the film plane. This is the important case for which the spin motion of the Fe ferromagnet creates spin dynamics in the nonmagnetic metal (here V) as discussed in the introduction. Commonly at a F-N interface there is also a magnetic moment induced in the N metal (e.g., Ref. 18). Unfortunately, in some of the literature dealing with transport mechanism and spin injection it is not mentioned. Both samples present a fourfold anisotropy within the film plane, the $[100]([110])$ being the easy (hard) in-plane direction and therefore we will present FMR results for these two principal directions, i.e., $\mathbf{H}\|[100]$ and $[110]$, respectively.

Figure 2 shows some experimental spectra. Due to a larger fourfold in-plane anisotropy of the Fe_4/V_2 SL the resonance positions of the two samples are slightly different.[14,17] The full resonance profiles as shown in Fig. 2 are analyzed. They show a perfect Lorentzian profile (first derivative $\partial\chi''/\partial H$). Nevertheless, we allow for our analysis of ΔH a sum of inhomogeneous ΔH_0 and homogeneous contributions $\Delta H_{\mathrm{Gilb}}+\Delta H_{2\mathrm{mag}}$,

$$\Delta H=\Delta H_0+\Delta H_{\mathrm{Gilb}}+\Delta H_{2\mathrm{mag}}. \tag{4}$$

The experimental result is shown in Fig. 3. Circles denote the Fe_4/V_4 sample and squares the Fe_4/V_2 sample. It is obviously clear that the Fe_4/V_2 sample shows a larger linewidth, but here it is important to measure down to 1 GHz to see the strong curvature in the datapoints and that at 1 GHz indeed the linewidth is very narrow, namely 30 Oe indicating again a high sample quality. It is also obvious at first glance that for both samples the full symbols measured along the $[100]$ direction show a larger linewidth than open symbols measured along the $[110]$ direction. In one case (open circles) data show an almost linear behavior (Gilbert damping is dominant). Quantitative systematic fits of the whole dataset leave almost no free parameters to play with: saturation magnetization was known to be $M_s=925$ G for Fe_4/V_4 and $M_s=1115$ G for Fe_4/V_2. The g values being isotropic in

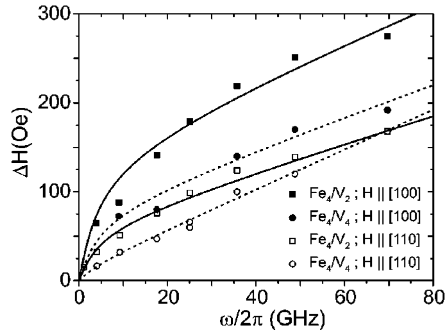

FIG. 3. Linewidth as a function of the microwave frequency $\omega/2\pi$ measured at room temperature for frequencies in the range 1–70 GHz. The squares indicate the $(Fe_4/V_2)_{60}$ SL, the circles denote the $(Fe_4/V_4)_{45}$ SL. The open symbols indicate the case with the external field parallel to the $[110]$ direction, whereas the filled symbols show the behavior for the external field applied parallel to the $[100]$ direction.

the film plane are $g=2.13$ forFe_4/V and $g=2.11$ in the case of Fe_4/V_2. The only open fit parameters were the Gilbert constant G [Eq. (2)] and the new two-magnon scattering strength Γ [Eq. (3)] plus a hypothetically inhomogeneous broadening ΔH_0. The latter turned out to be within experimental error bars zero $\Delta H_0=\pm 2$ Oe, proving again the high quality of our samples. We will come back later to the apparent ΔH_0^*. Γ and G are given in Table I. Several results can be deduced from the table.

(i) The linear Gilbert-damping constant G results very close to the bulk value. This is by no means obvious since we have used G as a free fit parameter to be optimized. For Fe G varies in the literature almost by a factor of 2, ranging from 0.58×10^8 s^{-1} to 1.3×10^8 s^{-1}.[19]

(ii) The new two-magnon process constant Γ is larger for the Fe_4/V_2 sample by a factor 2 to 3 than for the Fe_4/V_4 sample.

(iii) For both samples Γ is larger along the $[100]$ direction with respect to the $[110]$ direction. This is well understandable because previous structural investigations by means of x-ray diffraction (XRD) have proven that the interface is smoother for the Fe_4/V_4 sample and rougher for the Fe_4/V_2 specimen, approximately by a factor of 2 to 3. Also the fourfold in-plane anisotropy of the Fe_4/V_2 sample is larger and this shows a larger value of Γ.

This large fourfold in-plane anisotropy follows from an extension of the discussion in Ref. 5, provided we accept the picture of interface defects set forth there: on the statistical average, these are rectangular islands with sides parallel, on

TABLE I. Values for Γ and G for the two Fe/V SL's along the $[100]$ and $[110]$ directions.

Sample	Orientation	Γ (Oe)	$G(10^8$ s$^{-1})$
Fe_4/V_2	$\mathbf{H}\|[100]$	110	1.25
Fe_4/V_4	$\mathbf{H}\|[100]$	61	0.90
Fe_4/V_2	$\mathbf{H}\|[110]$	49	0.92
Fe_4/V_4	$\mathbf{H}\|[110]$	11	1.15

average, to [100] directions, as illustrated in Fig. 3 of Ref. 5. Suppose we now imagine that the magnetization makes an angle Θ with respect to [100] and note that the dominant contribution to the linewidth in Eq. (92) of Ref. 5 is proportional to the defect form-factor combination $(f_z - f_x)$. While only the case $\Theta = 0$ is examined in Ref. 5, extension of the calculation to general Θ shows that this form-factor combination has the angular variation $\cos(2\Theta)$. If the dominant term only is retained in the linewidth, and the defects are indeed rectangular as in the idealized picture in Ref. 5, then the two-magnon contribution to the linewidth will have the angular dependence $\cos(2\Theta)^2$, to actually vanish along the [110] direction. Of course, in any real sample, the defects will not have such a simple topology, so the angular variation will be less dramatic. However, the mechanism will lead to a fourfold in-plane anisotropy, and if the interface defects are oriented predominantly along [100], there will indeed be a fourfold anisotropy as we observe.

To visualize the essential importance of measurements over a large ω range we refer to our previous investigation of FMR linewidths.[20] There we measured only the low-frequency range and assumed like standard literature a linear Gilbert damping. As a result one gets too large apparent G values. In the other limit, if one measures only between 10 and 30 GHz, Fig. 1 illustrates clearly that this results in a nonreal residual linewidth ΔH_0^*.

In summary, clear experimental evidence is given that the linewidth of the FMR in ultrathin ferromagnetic interfaces is not exclusively controlled by the Gilbert damping. The successful phenomenological ansatz of the Gilbert damping in bulk ferromagnets can be seen as a ''viscosity type'' of damping in which the energy stored in the spin dynamics leaks into the thermal bath via spin-orbit interaction.[21] For ultrathin F-N interfaces we show that it is equally important to take into consideration the so-called two-magnon scattering at which the energy stored in the uniform motion of **M** scatters into spinwaves[22] of equal energy with $\mathbf{k} \neq 0$.[5] Depending on interface roughness and in-plane anisotropy field the two-magnon processes can easily reach scattering rates of 10^9 s^{-1}. Our experimental results in Fig. 3 having large and small contributions of the second type, respectively, demonstrate nicely that by manipulating the F-N interfaces on an atomic scale one might get into a position to change the spin damping and spin injection for currents perpendicular to the layer structure with the magnetization in-plane.

P. Blomquist and R. Wäppling are acknowledged for providing the samples and the XRD data. Fruitful discussions with B. Heinrich are acknowledged. This work was supported by the DFG Sfb290, TP A2.

---

*Corresponding author. FAX: +49-30-838-53646. Email address: babgroup@physik.fu-berlin.de

[1] Y. Li and K. Baberschke, Phys. Rev. Lett. **68**, 1208 (1992); Z.Q. Qiu, J. Pearson, and S.D. Bader, Phys. Rev. B **49**, 8797 (1994).

[2] M. Bander and D.L. Mills, Phys. Rev. B **38**, 12 015 (1988); R.P. Erickson and D.L. Mills, *ibid.* **43**, 11 527 (1991).

[3] P. Grünberg, J. Magn. Magn. Mater. **226–230**, 1688 (2001).

[4] R.E. Camley, J. Kwo, M. Hong, and C.L. Chien, Phys. Rev. Lett. **64**, 2703 (1990); R.W. Wang, D.L. Mills, Eric E. Fullerton, J.E. Mattson, and S.D. Bader, *ibid.* **72**, 920 (1994).

[5] R. Arias and D.L. Mills, Phys. Rev. B **60**, 7395 (1999); J. Appl. Phys. **87**, 5455 (2000).

[6] L. Berger, Phys. Rev. B **54**, 9353 (1996); J.C. Slonczewski, J. Magn. Magn. Mater. **159**, L1 (1996); Y. Tserkovnyak, A. Brataas, and G.E.W. Bauer, Phys. Rev. Lett. **88**, 117601 (2002); R. Urban, G. Woltersdorf, and B. Heinrich, *ibid.* **87**, 217204 (2001).

[7] L. Landau and E. Lifshitz, Phys. Z. Sowjetunion **8**, 153 (1935).

[8] T.L. Gilbert, Phys. Rev. **100**, 1243 (1955).

[9] Z. Celinsky and B. Heinrich, J. Appl. Phys. **70**, 5935 (1991).

[10] N. Bloembergen, Phys. Rev. **78**, 572 (1950).

[11] V. Kambersky and C.E. Patton, Phys. Rev. B **11**, 2668 (1975); M.J. Hurben and C.E. Patton, J. Appl. Phys. **83**, 4344 (1998).

[12] B. Heinrich, J.F. Cochran, and R. Hasegawa, J. Appl. Phys. **57**, 3690 (1985); R. Urban, B. Heinrich, G. Woltersdorf, K. Ajdari, K. Myrtle, J.F. Cochran, and E. Rozenberg, Phys. Rev. B **65**, 020402(R) (2001).

[13] P. Isberg, B. Hjörvarsson, R. Wäppling, E.B. Svedberg, and L. Hultman, Vacuum **48**, 483 (1997).

[14] A.N. Anisimov, W. Platow, P. Poulopoulos, W. Wisny, M. Farle, K. Baberschke, P. Isberg, B. Hjörvarsson, and R. Wäppling, J. Phys.: Condens. Matter **9**, 10 581 (1997).

[15] A. Broddefalk, R. Mathieu, P. Nordblad, P. Blomqvist, R. Wäppling, J. Lu, and E. Olsson, Phys. Rev. B **65**, 214430 (2002).

[16] D. Labergerie, C. Sutter, H. Zabel, and B. Hjörvarsson, J. Magn. Magn. Mater. **192**, 238 (1999).

[17] A.N. Anisimov, M. Farle, P. Poulopoulos, W. Platow, K. Baberschke, P. Isberg, R. Wäppling, A.M.N. Niklasson, and O. Eriksson, Phys. Rev. Lett. **82**, 2390 (1999).

[18] A. Scherz, H. Wende, P. Poulopoulos, J. Lindner, K. Baberschke, P. Blomqvist, R. Wäppling, F. Wilhelm, and N.B. Brookes, Phys. Rev. B **64**, 180407(R) (2001).

[19] M.B. Stearns, in *Magnetic Properties of Metals*, edited by H.P.J. Wijn, Landolt-Börnstein, New Series, Group III, Vol. 19, Pt. a (Springer, Berlin, 1986).

[20] W. Platow, A.N. Anisimov, G.L. Dunifer, M. Farle, and K. Baberschke, Phys. Rev. B **58**, 5611 (1998).

[21] After first submission of our manuscript the recent theoretical work by A.Yu. Dobin and R.H. Victora, Phys. Rev. Lett. **90**, 167203 (2003) appeared. This also discusses the nonlinear FM relaxation and its general importance for spin dynamics. The present work gives experimental evidence for this and is interpreted using the nonlinear two-magnon mechanism.

[22] Harry Suhl, IEEE Trans. Magn. **34**, 1834 (1998).