Spin-freezing transition in a CoO/Permalloy bilayer revealed by transverse ac susceptibility

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We utilize variable-temperature, variable-frequency magneto-optical transverse magnetic susceptibility technique to study the static and dynamical magnetic properties of a thin-film CoO/Permalloy bilayer. Our measurements demonstrate that the exchange bias - the asymmetry of the hysteresis loop - is caused not by the unidirectional anisotropy, but rather by the difference between the energy barriers between two reversed states of magnetization stabilized by a large uniaxial anisotropy. We also observe abrupt variation of the frequency-dependent imaginary part of ac susceptibility near the exchange bias blocking temperature, consistent with the spin glass transition close to this temperature. Our results suggest a route for the precise characterization and control of the dynamical and static characteristics of thin-film magnetic heterostructures, with possible applications in reconfigurable magnonic and neuromorphic circuits.

I. INTRODUCTION

Intense ongoing research in magnetism is motivated by a variety of phenomena resulting from the strong coupling of spin to structural, optical, and electronic properties of matter, with promising applications in sensing and information technology.[1] Historically, the main focus has been on the ferromagnetically (F) and ferrimagnetically ordered materials, thanks to their robust magnetic ordering that can be efficiently manipulated by the magnetic fields or spin currents, and detected by a variety of electronic and optical techniques.[2] Other types of magnetic ordering have recently attracted a significant attention, due to the possible advantages they can provide in downscaleing and increased speed of magnetic nanodevices. For instance, the burgeoning field of antiferromagnetic (AF) spintronics is motivated by the considerably faster than in the ferromagnets timescales of magnetization dynamics, and their negligible susceptibility to perturbing magnetic fields.[3] Frustrated magnetic systems forming spin liquids have also attracted significant attention, thanks to the rich physics associated with exotic states such as fractionalized excitations that can be hosted by such systems.[4]

Magnetic heterostructures composed of materials with different magnetic properties provide an efficient approach to engineering the magnetic and electronic properties of thin films. Some of the classic examples include the exchange bias (EB) effect observed in F/AF bilayers[5] and giant magnetoresistance in magnetic spin valve heterostructures.[6] One of the general questions arising in this context concerns the nature of magnetism in heterostructures of thin films with different magnetic properties. In case of F/AF bilayers, the magnetic ordering of the two materials is incompatible: with a rare exception of atomically flat uncompensated surfaces of AF, the magnetic energies of both F and AF generally cannot be simultaneously minimized in their ordered state, due to the frustrated exchange interaction at their interface.[6] This can lead to the emergence of new magnetization states distinct from the F or AF ordering of individual layers. In thick films, the modest effects of interfacial effective exchange fields result in the formation of multidomain states of AF and/or F.[8] The characteristic size \( l \) of the domains is expected to decrease with increasing effective interfacial exchange field, which for thin films scales inversely with the film thickness. For sufficiently small film thicknesses, \( l \) becomes smaller than the domain wall width \( \delta \) determined by the magnetic anisotropy of the material. The magnetization no longer forms distinct domains separated by domain walls, and a new “Heisenberg domain” state is expected to emerge.[9]

The existence of the Heisenberg domain state is still experimentally unverified, and its dynamic and thermodynamic properties remain unknown. Recent magneto-electronic measurements of the time-dependent magnetization state for three common AFs - FeMn, CoO, and NiO - have demonstrated universal slow power-law aging at low temperatures.[10-12] Aging was observed only in thin films, suggesting that it is likely closely related to the emergence of the Heisenberg domain state. The dependence of aging on the magnetic history was found to be inconsistent with the Arrhenius-type activation, indicating cooperative behaviors akin to the avalanche dynamics in spin glasses.[13] The relationship between the Heisenberg domain state and spin glasses was further elucidated by the temperature dependence of aging in thin NiO films.[13] Aging was slow and independent of temperature \( T \) below the exchange bias blocking temperature \( T_B \), but abruptly accelerated above \( T_B \). This observation, together with the analysis of the dependence on the magnetic history, was interpreted in terms of the spin glass transition from a fragile spin-solid at \( T < T_B \) to a correlated spin-liquid at \( T > T_B \). These results suggested the possibility to engineer magnetism in thin films, which can become useful for new applications in information technology. However, time-domain aging measurements did not provide sufficient information for the quantitative understanding of these systems.

Here, we show that a significant insight into the nature of the magnetization states in thin F/AF films can be provided by the transverse ac susceptibility tech-
which enables phase-sensitive characterization of the dynamical magnetization response. We utilize this technique to show that at temperatures above the Neel temperature $T_N$ of AF=CoO, the susceptibility is real and independent of frequency, and is identical to that of a standalone ferromagnet. The susceptibility is also real at temperatures far below $T_B$, but is significantly smaller than that of a single ferromagnet, as expected due to the effects of exchange interaction between F and the spin-solid state of AF.

Our central result is the observation of a large and strongly temperature- and frequency-dependent imaginary part of susceptibility near $T_B$, associated with the viscous losses in AF. Analysis shows that the magnetic viscosity varies by four orders of magnitude over the temperature range of 20 K, which in the standard Nee-Brown theory of magnetic viscosity would require unphysically large activation attempt rates. This indicates that a spin-freezing transition occurs near $T_B$, supporting the conjecture that the Heisenberg domain state is a spin glass. In addition, the ability to precisely characterize the magnetic anisotropy allowed us to establish that in the studied system, the asymmetry of the hysteresis loop is not caused by the unidirectional anisotropy commonly identified with EB, but rather by the different reversal energy barriers between the opposite magnetization states stabilized by the uniaxial anisotropy. These results shed light on the exchange interactions at F/AF interfaces, and their effects on the static and dynamical characteristics of magnetization states.

II. EXPERIMENT AND SAMPLE DETAILS

In this Section, we outline the experimental technique, describe the setup utilized in our measurements, and provide details on the sample preparation.

II.1. Transverse magnetic ac susceptibility

Measurements of ac susceptibility have been widely utilized for the characterization of phases of matter. For instance, the low-frequency divergence of susceptibility provides a robust indication for second-order phase transitions, while its dependencies on temperature, frequency and other experimental parameters have been utilized to establish the universal laws governing these transitions. The frequency-dependent peak observed in ac susceptibility at the glass transition temperature has been extensively studied as one of the key signatures of the underlying phenomena. It has been also suggested that glass transition may be associated with a weak logarithmic low-frequency divergence of susceptibility, which may become important for the identification and classification of frustrated states of matter.

Magnetic susceptibility generally depends on the direction of the driving ac field. In the studies of F/AF bilayers such as those described below, the F layer becomes saturated at modest $H_{dc}$, while direct coupling of the Neel order of AF to $H_{dc}$ is negligible. In this configuration, a small ac field $h_{ac} = H_{ac} \cos \omega t$ applied parallel to H does not significantly perturb the magnetic system. Here and throughout this paper, we assume that the frequency $\omega = 2\pi$ is significantly smaller than the characteristic dynamical frequencies of the magnetic system, such as the ferromagnetic or parametric resonances of F. For $H_{ac}$ transverse to H, the magnetization $M$ of F is expected to oscillate, following the direction of the total field $H_{dc} + H_{ac}$. The transverse magnetic susceptibility can be defined as $\chi_T = M_T/H_{ac}$, where $M_T$ is the oscillating component of $M$ transverse to the dc field. This quantity does not directly reflect the response of AF to perturbations, which is the main subject of the present work, since it remains finite even in the absence of AF. Therefore, to avoid confusion about the response studied in our work, in this paper we focus on the measurements and analysis of ac magnetization $M_T$.

The magnetization $M$ of F experiences not only external magnetic fields, but also exchange interactions at the F/AF interface. By analyzing the oscillation of $M$ in response to $H_{ac}$, one can characterize this interaction, as follows. Assume that the effect of AF on $M$ can be described by time-independent effective unidirectional anisotropy field $H_{ud}$. We can define the total effective static field $H' = H_{dc} + H_{ud}$, such that $M$ follows the instantaneous direction of the total effective field $H' + H_{ac}$. Fig. (a). The normalized ac magnetization $M_T/M$ is then given by

$$\frac{M_T}{M} = \frac{1}{\sqrt{1 + (H'/H_{ac})^2}}. \tag{1}$$
optical cryostat. Planar magneto-optical Kerr effect 

−susceptibility measurement, performed in a 80 transition.

imize the inhomogeneous broadening of the spin-freezing rms, as measured by the atomic force microscopy, to min-

larger than the typical CoO roughness of about 0

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bias. The CoO(1.5) insert between CoO and Py served as an oxygen 

diffusion barrier preventing the oxidation of Py, which 

Co(1.5)Py(50)AlOx(2) multilayers were de-

posited at room temperature on the oxidized Si substrate 

by magnetron sputtering, in an ultrahigh vacuum chamber 

with the base pressure of 7 × 10−9 Torr. All thicknesses 

are given in nanometers. The deposition rates varied from 0.2 A/s for AlOx to 1.8 A/s for Py, and were 

calibrated using a quartz crystal microbalance. The deposition was performed in an in-plane magnetic field of 

about 100 Oe applied in the direction perpendicular to the dc field used in our measurements, to facilitate the establishment of exchange bias. The CoO layer was sputtered in Ar/O2 mixture, using the deposition parameters optimized in our previous studies of CoO-based heterostructures. The other materials were sputtered from the stoichiometric targets in ultrahigh purity Ar. The soft ferromagnetic Py was chosen to minimize the effects of anisotropy on the magnetic susceptibility. The Co(1.5) insert between CoO and Py served as an oxygen diffusion barrier preventing the oxidation of Py, which in our past experience resulted in a gradual deterioration of the magnetic characteristics related to exchange bias. The optically transparent AlOx capping layer was utilized to protect the surface of Py from oxidation.

The thickness of x = 6 nm for the CoO layer, selected for the measurements described below, was sufficiently small so that this material was expected to form the Heisenberg domain state, as inferred from the prior time-domain studies of thickness-dependent magnetization aging. On the other hand, this thickness was significantly larger than the typical CoO roughness of about 0.3 nm rms, as measured by the atomic force microscopy, to minimize the inhomogeneous broadening of the spin-freezing transition.

II.2. Sample

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posited at room temperature on the oxidized Si substrate by magnetron sputtering, in an ultrahigh vacuum chamber with the base pressure of 7 × 10−9 Torr. All thicknesses are given in nanometers. The deposition rates varied from 0.2 A/s for AlOx to 1.8 A/s for Py, and were calibrated using a quartz crystal microbalance. The deposition was performed in an in-plane magnetic field of about 100 Oe applied in the direction perpendicular to the dc field used in our measurements, to facilitate the establishment of exchange bias. The CoO layer was sputtered in Ar/O2 mixture, using the deposition parameters optimized in our previous studies of CoO-based heterostructures. The other materials were sputtered from the stoichiometric targets in ultrahigh purity Ar. The soft ferromagnetic Py was chosen to minimize the effects of anisotropy on the magnetic susceptibility. The Co(1.5) insert between CoO and Py served as an oxygen diffusion barrier preventing the oxidation of Py, which in our past experience resulted in a gradual deterioration of the magnetic characteristics related to exchange bias. The optically transparent AlOx capping layer was utilized to protect the surface of Py from oxidation.

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II.3. Measurement technique

Figure 1(b) shows a schematic of our transverse ac susceptibility measurement, performed in a 80 − 325 K optical cryostat. Planar magneto-optical Kerr effect (MOKE) was used to detect the dynamical magnetization of the sample. An external electromagnet produced an in-plane dc magnetic field \( H_{dc} \), of up to 1 kOe, while the in-plane ac field \( H_{ac} \) transverse to the dc field was produced by a coil wrapped on a slotted toroidal ferrite core built into the cryostat. The sample was mounted on a silicon cold finger, with the Cernox thermometer mounted within less than 1 mm from the sample. The slotted ferrite core of the ac coil confined the ac magnetic field, minimizing the parasitic effects of induced emf in the metallic parts of the cryostat, as well as the Faraday effect in the optical cryostat window. At room temperature \( T = 295 \) K, this setup allowed us to achieve ac fields of more than 500 Oe at frequencies \( f \) of a few Hz, and over 100 Oe at the maximum measurement frequency \( f = 7 \) kHz. The maximum achievable ac field was somewhat reduced at lower \( T \), due to the decreased magnetic permeability of ferrite. The amplitude and the phase of the field produced by the coil were calibrated to the precision of ±2% over the temperature and frequency ranges of our measurements, by utilizing a 50 nm-thick Py film whose response followed Eq. (1) with \( H' = H_{dc} \).

Magnetic-optical measurement of the dynamical magnetization state utilized a polarized HeNe laser spot at a 60° angle of incidence. The polarization rotation resulting from the planar MOKE effect was periodically modulated by the oscillating magnetization. This rotation was converted into the intensity oscillations by an analyzer. The oscillations of photocurrent produced by the Si photodetector were measured by the phase-sensitive lockin amplifier. We note that the longitudinal MOKE effect, determined by the projection of magnetization on the dc field, may also contribute to the polarization rotation, but by the symmetry of our measurement geometry, this effect produced only 2\( f \) and higher harmonics that were rejected by the lockin.

III. EXPERIMENTAL RESULTS

III.1. Transverse ac susceptibility at \( T > T_N \)

At temperatures \( T \) above the Neel temperature \( T_N = 291 \) K of CoO, this layer is not expected to affect the transverse susceptibility of Py, allowing us to validate the transverse ac susceptibility technique, and establish its effectiveness in the quantitative characterization of magnetic properties. Figure 2 shows the results for \( T = 320 \) K. The magnetic hysteresis loop, obtained using a vibrating sample magnetometer, is characterized by the coercivity of 3 Oe, and the saturation field of about 15 Oe [Fig. 2(a)]. The normalized real part of ac magnetization \( R e((M_T/\mathcal{M}) \) rapidly increases with decreasing magnitude of \( H_{dc} \), peaks at \( H_{dc} = \pm 16 \) Oe, and reduces to zero at \( H_{dc} = 0 \). Meanwhile, the imaginary part \( I m((M_T/\mathcal{M}) \) remains negligible at all \( H_{dc} \), except for the sharp peaks at \( H_{dc} = \pm 16 \) Oe coinciding with the peaks of the real part. These data were acquired at \( f = 1 \) kHz; the results were
Figure 2. (Color online). Results of measurements at $T = 320$ K, above the Neel temperature of CoO. (a) Magnetic hysteresis loop with in-plane field. (b) Real and imaginary parts of normalized ac magnetization vs field, as labeled. Note that the value of the real part for the upward field scan (curve) and for the downward scan (symbols) coincide. Measurements were performed at $H_{ac} = 5$ Oe, $f = 1$ kHz. (c) Symbols: inverse of Re($M_{dc}/M$) from panel (b), lines: linear fits. The small-$H$ data, which are off this plot’s scale, were removed. (d) Real (open symbols) and imaginary (solid symbols) parts of normalized transverse magnetization vs ac field, at $H_{dc} = 100$ Oe, $f = 1$ kHz. Curve is a fit with Eq. (1), yielding $H' = 70.5 \pm 1$ Oe. The normalization in panels (b)-(d), determined from the fitting with Eq. (1) was consistent among different data sets.

independent of $f$ over the accessible range. The susceptibility does not exhibit any noticeable hysteresis in the field dependence. Therefore, the peaks at $H_{dc} = \pm 16$ Oe cannot be attributed to the hysteretic magnetization reversal. The inverse of Re($M_{dc}/M$) is well approximated by two linear dependencies, intercepting the field-axis at 11 Oe for $H_{dc} > 0$, and −11 Oe for $H_{dc} < 0$. According to Eq. (2), the effective exchange-bias field $H_{ud}$ would have produced the same intercept for both field directions, rather two opposite offsets.

The observed behaviors can be explained by the small uniaxial anisotropy, acquired by Py due to the in-field deposition, with the easy axis perpendicular to $H_{dc}$. Indeed, in the absence of ac field, we can write the magnetic energy density of Py as

$$u = -M H_{dc} \cos(\theta) - M H_{ua} \cos^2(\theta)/2,$$

where $\theta$ is the angle formed by the Py magnetization $\mathbf{M}$ relative to the dc field [see Fig. 1(a)], and $H_{ua}$ is the effective uniaxial anisotropy field, defined to be positive for the easy axis parallel to $H_{dc}$. For small $\theta$, $u \approx -M H_{dc} \theta^2/2 - M H_{ua} \theta^2/2 + \text{const}$. The effect of uniaxial anisotropy transverse to $H_{dc}$ is equivalent to that of the field $H_{ua}$ for $H_{dc} > 0$, and $-H_{ua}$ for $H_{dc} < 0$. The response to $H_{ac}$, which is determined by the derivative $\partial u/\partial \theta (\theta = 0)$, vanishes at $H_{dc} = -H_{ua}$, resulting in the peaks in Fig. 2(b), where $H_{ua} = -11$ Oe is opposite to $H_{dc}$ for the easy anisotropy axis perpendicular to $H_{dc}$.

Unavoidable local variations of anisotropy result in an inhomogeneous magnetization state at $H_{dc}$ close to $H_{ua}$, giving rise to dynamical losses manifested as the peaks of Im($M_{dc}/M$). At $H_{dc} = 0$, the magnetization is expected to rotate to the easy-axis direction, perpendicular to the dc field and parallel to the ac field. As discussed above, ac susceptibility is then expected to vanish, consistent with the data in Fig. 2(b). Finally, uniaxial anisotropy also explains the tilting of the hysteresis loop Fig. 2(a), resulting in a relatively large, compared to the typical values for Py, saturation field.

We note that by fitting $M_{dc}/M$ vs $H$ with Eq. (2) separately for $H > 0$ and $H < 0$, one can obtain both the directional and the uniaxial anisotropies, according to

$$H_{ua} = (H_+ - H_-)/2, \quad H_{ud} = -(H_+ + H_-)/2,$$

where $H_+$, $H_-$ are the intercepts of the linear fits for $H_{dc} > 0$ and $H_{dc} < 0$, respectively. If $H_+$ and $H_-$ were positive and negative coercive fields, respectively, then Eqs. (3) would become identical, up to a sign, to the definitions of the coercive field $H_C$ and the exchange bias field $H_E$ commonly introduced in the studies of exchange bias. These two sets of characteristics are not directly related, as shown below.

A complementary approach to analyzing the magnetic properties is provided by the nonlinear regime of Eq. (1), as follows. Consider the dependence of $M_{dc}/M$ on $H_{ac}$. According to Eq. (2), this dependence is linear at $H_{ac} \ll H'$, with the slope $1/H'$. This slope cannot be used to independently determine $H'$, because the overall scaling of the measurement must be determined from the data such as Fig. 2(c). However, at $H_{ac}$ comparable to $H'$, the dependence becomes nonlinear. Keeping two leading-order terms in the expansion of Eq. (2) with respect to $H_{ac}/H'$, we obtain

$$M_{dc}/M \approx H_{ac}/H' - 1/2 \left( H_{ac}/H' \right)^3.$$

This equation describes curving down due to the saturation of $M_{dc}$, when the oscillating magnetization approaches the limiting value of $M$ at large $H_{ac}$. The ratio of the cubic to the linear terms in Eq. (5) provides a quantitative measure of $H_{ac}/H'$, regardless of the overall data scaling, and independent from the results of linear-regime fitting discussed above. More generally, fitting of the nonlinear data with Eq. (1), with the overall data scale used as an adjustable parameter, provides an independent characterization of $H'$.

Figure 2(d) shows $M_{dc}/M$, measured with $H_{ac}$ varied from 0 to 200 Oe, at $H_{dc} = 100$ Oe. The imaginary part of ac magnetization is negligible, as expected for the saturated state at $H > H_{ua}$. The real part linearly
increases at small $H_{ac}$, but at large $H_{ac}$ starts to saturate towards 1, in agreement with our analysis. The dependence Eq. [1] provides a good fitting [curve in Fig. 2(d)], slightly overshooting the data at $H_{ac}$ 100 Oe, and undershooting near 200 Oe.

The value $H' = 70.5 \pm 1$ Oe, extracted from the fitting, seems to imply that the effective anisotropy field is three times larger than the value obtained from fitting $M_T(H_{dc})$. However, the concept of the effective anisotropy field, based on the small-angle expansion of Eq. [3], is not applicable to the large-amplitude magnetization oscillation. To determine the actual dependence of oscillation amplitude on field, one must minimize the full energy functional Eq. (3), with an additional Zeeman term describing the ac field. Quantitative analysis of this dependence is beyond the scope of the present work. Qualitatively, one can expect that the torque exerted on the magnetization due to the uniaxial anisotropy is reduced when the magnetization rotates close to the easy anisotropy axis. In contrast, in the effective field approximation, this torque reaches a maximum at angle $\theta_F = \pi$. Thus, the effective-field approximation is expected to underestimate the amplitude at large $H_{ac}$. Accordingly, the effective-field approximation Eq. 5 understimates the curving, resulting in the overestimation of $H_{ua}$.

III.2. Transverse ac susceptibility at $T < T_N$

The results for $T > T_N$ presented above confirm that the transverse ac susceptibility technique enables precise characterization of anisotropy, by analyzing its dependencies on the dc and ac fields. Here, we show how this approach can be utilized to gain insight into the nature of the magnetization states in F/AF bilayers below the Neel temperature of AF, $T_N = 291$ K for CoO. In all of the measurements described below, the sample was cooled from $T = 320$ K at $H_{dc} = 500$ Oe, at a rate of about 0.5 K/s.

We start with the simpler behaviors observed at sufficiently low temperatures. Figure 3(a) shows a representative $M_T/M$ vs $H_{dc}$ dependence for $T = 80$ K. The overall trends are qualitatively similar to the high-temperature results shown in Fig. 2(b), with a notable exception of asymmetry with respect to the field direction, as expected due to EB. The values of the reversal fields, determined from these data, are $H_1 = -40$ Oe and $H_2 = -450$ Oe for the upward and downward field sweeps, respectively, yielding the effective EB field $H_E = -(H_1 + H_2)/2 = 245$ Oe and coercivity $H_C = (H_1 - H_2)/2 = 205$ Oe.

In the studies of EB, asymmetry between the reversal fields $H_1$, $H_2$, the dependence $M_T(H_{dc})$ is almost symmetric with respect to the field reversal, implying that $H_{ud}$ is small [see Eq. (4)]. Indeed, fitting with Eq. (2) [curves in Fig. 3(a)] yields $H_{ud} = 47 \pm 1$ Oe, five times smaller than $H_E$. Meanwhile, $H_{ua} = 428 \pm 1$ Oe [easy axis along $H_{dc}$] is larger than $H_{ud}$ by an order of magnitude, and is twice as large as $H_C$. A discrepancy between unidirectional anisotropy and asymmetry of magnetization reversal has been reported for other F/AF systems.[27]

The origin of this discrepancy is elucidated by the detailed inspection of Fig. 3(a). In the downward field sweep, $M_T/M$ grows to 0.12 at $H = -320$ Oe, before rapidly decreasing due to the magnetization reversal. This dependence is well fitted by Eq. (2) [curve in Fig. 3(a)], demonstrating that the magnetization remains in a uniform state opposing the dc field, until the effective exchange field is almost completely compensated by $H_{dc}$. In contrast, in the upward field sweep, the reversal occurs at the point of intersection between the positive- and negative-field dependencies, corresponding to a significantly smaller $M_T/M = 0.06$. Thus, the reversal occurs as soon as the effective field in the reversed direction becomes larger than that in the forward direction. We can conclude that in the studied system, the asymmetry between the reversal fields is associated almost entirely with the different energy barriers for the magnetization reversal in the opposite directions.

Since the variations of $M_T/M$ in Fig. 3(a) are well approximated by Eq. 2, and $\text{Im}(M_T/M)$ is negligible, aside from a sharp peak accompanying the downward-
sweep reversal, the nonuniform magnetization states of Py do not play a significant role in the observed behaviors. Therefore, the observed asymmetry of magnetization reversal mechanisms cannot be attributed to the nonuniform spatial distribution of the unidirectional anisotropy direction/magnitude. We speculate that the observed behaviors are associated with canting (or spin-flop) of the AF magnetic sublattices towards $M$, due to the large exchange interaction at F/AF interface. Such canting, with the Neel order transverse to $M$, was proposed as the mechanism of EB canting, with the Neel order transverse to $M$ at the large exchange interaction at F/AF interface. Such characteristics, determined from the hysteresis loops of change bias in the studied CoO/Py bilayer is confirmed in F/AF bilayers. Our data reconcile these seemingly contradictory theoretical results: the dominant effect is uniaxial anisotropy, likely associated with the reversible canting of AF sublattices, while the asymmetry of magnetization reversal is caused by the different energy barriers for the reversal of the AF canting direction. This result is likely related to the prior observations of asymmetric spatial characteristics of magnetization reversal in F/AF bilayers. Our interpretation is also consistent with experimental observations of uncompensated AF spins at F/AF interfaces, which showed that most of these spins reverse together with $M$, while only a small percentage responsible for the unidirectional anisotropy remain fixed.

The irrelevance of unidirectional anisotropy to exchange bias in the studied CoO/Py bilayer is confirmed by the temperature dependence of the magnetic characteristics, determined from the hysteresis loops of $M_T/M$ acquired at temperatures between 80 K and 320 K, Fig. 3(b). The coercivity $H_C$, the exchange bias field $H_E$, and the effective uniaxial anisotropy field $H_{ua}$ monotonically decrease with increasing $T$, Fig. 3(b). The exchange bias vanishes above 220 Oe, but the enhanced coercivity and uniaxial anisotropy persist up to 240–250 K. Surprisingly, the unidirectional anisotropy $H_{ud}$ remains small and almost constant up to 220 K, with a small bump around 120 K. These results clearly demonstrate that $H_E$ is not correlated with the unidirectional anisotropy, but follows the uniaxial anisotropy, consistent with the analysis above.

We now discuss the behaviors of ac susceptibility close to the exchange bias blocking. In this temperature range, the ac magnetization exhibits a sizable imaginary part, as illustrated in Fig. 3(c) for $T = 240$ K. The variations of $\text{Im}(M_T/M)$ in Fig. 3(c) appear to simply mirror $\text{Re}(M_T/M)$, but the relation between these two parts is more complicated, as shown by the dependence on the ac field in Fig. 3(d). While $\text{Re}(M_T/M)$ monotonically increases with increasing $H_{ac}$, the magnitude of the imaginary part exhibits a shallow maximum at $H_{ac} = 130$ Oe.

Since $\text{Im}(M_T)$ does not vanish at large $H_{ac}$, the large imaginary part cannot be attributed to the losses associated with the inhomogeneity of $M$. Instead, it likely originates from the viscous dynamics of AF magnetization previously observed as rotatable AF-induced anisotropy producing a dynamical effective exchange field, exerted on $M$, whose phase lags behind that of $H_{ac}$. The remainder of this paper is focused on characterizing this dynamics and analyzing its implications for the nature of the magnetization state of AF.

III.3. Dependence of transverse ac susceptibility on frequency

The dynamics of AF magnetization violates the approximations underlying Eq. (1), so the response is expected to be generally frequency-dependent. We have utilized the broadband capability of our measurement setup to analyze this dependence in the range of frequencies from a few Hz to several kHz. On the low-frequency side, this range is limited by the $1/f$ noise, and on the high-frequency side, it is limited by the properties of the ferrite core in the ac coil in our setup, whose permeability is reduced at high frequencies, especially at low temperatures.

In measurements of the dependence on the dc field near $T_B$, ac magnetization depended on the magnetic history and evolved in time, due to the previously demonstrated magnetic aging. To avoid this complication, we focus on measurements performed at a constant $H_{dc}$, after a sufficiently long delay to minimize the effects of aging. The dependence of $\text{Re}(M_T/M)$ on $H_{ac}$ was well described...
by Eq. (1) over the entire studied ranges of temperatures and frequencies [see Fig. 3(d)], allowing us to determine the effective field \( H' \). In all these measurements, the imaginary part of ac magnetization never exceeded 15% of the real part, making the distinction between the real part and the total ac magnetization insignificant.

Figure 4(a) shows the dependence of \( H' \) on frequency for four representative values of \( T \), at \( H_{dc} = 100 \) Oe. At \( T = 320 \) K above \( T_N \), \( H' = 71 \pm 1 \) Oe is constant over the range \( f = 1 - 1000 \) Hz, except for a single outlier \( H' = 68 \pm 2 \) Oe at \( f = 1 \) Hz. Below \( T_N \), the effective field is close to its high-temperature value at small \( f \), and increases with increasing frequency [see the results for \( T = 240 \) K in Fig. 4]. This behavior is indicative of viscous AF magnetization dynamics, whose effects disappear in the low-frequency limit. At temperatures near \( T_B \), the dependence of \( H' \) on \( f \) becomes significant, and at low frequencies it no longer approaches the high-temperature value. For instance, at \( T = 215 \) K, \( H' \) varies from 192 Oe at 1 Hz to 325 Oe at 800 Oe. We note that this variation is too large to be associated with the unidirectional anisotropy [see Fig. 3(b)]. Instead, it likely originates from the dynamics of the canted AF magnetization, which underlies the uniaxial anisotropy of ferromagnets. As the temperature is further reduced, \( H' \) becomes large and independent of \( f \), as illustrated in Fig. 3(a) for \( T = 200 \) K. This behavior is consistent with the spin-solid state of AF magnetization, providing a frequency-independent elastic contribution to \( H' \).

Note that significant variations of frequency-dependent effective exchange field occur within a relatively narrow range of temperatures near \( T_B \), suggesting that the AF magnetization dynamics may exhibit a critical slowdown. This hypothesis is supported by measurements of the dependence of ac magnetization on frequency at constant dc and ac fields, Figs. 3(b),(c). The real part is independent of frequency at temperatures above \( T_N \) and far below \( T_B \). As the temperature is decreased below \( T_N \) but above \( T_B \), \( Re(M_{f}/M) \) starts to decrease with increasing frequency, while still approaching its high-temperature value at small \( f \) [see the \( T = 250 \) K data in Fig. 3(b)]. The variation is most significant near \( T_B \), where it is well described by the logarithmic function over the experimentally accessible frequency range. As the temperature is further reduced, the variation of \( Re(M_{f}/M) \) with \( f \) rapidly diminishes. These behaviors are consistent with the interpretation discussed above: negligible effects of AF at high temperatures, elastic spin solid state at sufficiently low temperatures, and viscous AF dynamics near \( T_B \).

The central result of this paper is the observation of a qualitative change, near \( T_B \), in the frequency dependence of the imaginary part of ac magnetization, providing strong evidence for the critical slowing down of AF dynamics. The value of \( Im(M_{f}/M) \) is negligible above \( T_N \) and far below \( T_B \), as expected from the discussion above. As the temperature is decreased below \( T_N \), an increasingly significant imaginary part emerges at high frequencies, whose magnitude monotonically decreases towards zero in the low-frequency limit, as shown for \( T = 250 \) K in Fig. 4(c). In contrast, at \( T = 240 \) K, \( Im(M_{f}/M) \) exhibits a broad peak around \( f = 100 \) Hz, while at \( T = 230 \) K the magnitude of \( Im(M_{f}/M) \) monotonically increases with decreasing \( f \). At lower \( T \), the slope of the dependence at high frequencies decreases, and the value of \( Im(M_{f}/M) \) decreases towards zero in the low-frequency limit, as illustrated in Fig. 3(c) for \( T = 215 \) K.

The abruptness of the variation of \( Im(M_{f}/M) \) near \( T_B \) is further illustrated by its dependence on temperature, Fig. 4(d). This dependence exhibits a sharp peak with the full width at half maximum of about 30 K, with the maximum at \( T = 240-250 \) K correlated with the onset of magnetic anisotropies associated with EB [see Fig. 3(b)]. The position of the peak does not noticeably depend on the measurement frequency, as shown for \( f = 100 \) Hz and 1 kHz. These observations cannot be explained by the thermal activation dynamics over a broad distribution of weakly temperature-dependent magnetic energy barriers, often invoked in the context of EB in monodisperse magnetic nanoparticles, i.e. it is too narrow for the Arrhenius activation.

IV. ANALYSIS

The main challenge in analyzing the transverse ac susceptibility of F/AF bilayers is associated with the indirect relationship between this quantity and the dynamical response of AF. As discussed in the Introduction. In particular, the susceptibility remains finite even if the effects of AF are negligible, or if it forms a rigid spin-solid. In Section 3, we described a static-AF approximation for the transverse ac susceptibility, which predicted that the ac magnetization oscillates in-phase with ac field, with the amplitude independent of frequency. As we have seen in Section 3.3, the most intriguing behaviors, observed near \( T_B \), are associated with the frequency-dependent susceptibility characterized by a significant imaginary part. To understand the implications of these

![Figure 5. (Color online). Schematic of the model for the ac transverse susceptibility incorporating the dynamical response of AF.](image-url)
observations for the dynamics of AF, we develop a model connecting them to the dynamical properties of AF.

Our model is illustrated in Fig. 5. The time dependence of the ac field is described by \( h_{ac}(t) = H_{ac}\cos(\omega t) \). The oscillation of the magnetization \( \mathbf{M} \) is described by the time-dependent angle \( \theta_F(t) = Re\theta_F\cos(\omega t) - Im\theta_F\sin(\omega t) \). The effect of exchange interaction at the F/AF interface can be described in terms of the time-dependent effective exchange field \( h_{AF} \) with magnitude \( H_{AF} \approx H_{ud} \). It is aligned with the AF magnetization, forming an angle \( \theta_{AF}(t) = Re\theta_{AF}\cos(\omega t) - Im\theta_{AF}\sin(\omega t) \) with respect to the dc field \( H_{dc} \). To keep our analysis tractable, we consider small-angle limit \( H_{ac} \ll H_{dc} \), such that \( \theta_F, \theta_{AF} \ll 1 \).

The elastic and the viscous contributions to the AF dynamics, driven by the exchange interaction with \( F \), can be described by

\[
K\theta_{AF} + \nu \frac{d\theta_{AF}}{dt} = \theta_F - \theta_{AF},
\]

where \( K \) is the elastic coefficient defined by the static magnetic properties of AF and the effective exchange field at the F/AF interface, and \( \nu \) is the effective viscosity in units of time, determined by the energy relaxation processes in AF. In the Neel-Brown theory, \( \nu \) would be the thermal activation time \( \tau_{th} \). The magnetization state of the ferromagnet is determined by the balance of the torques exerted on \( \mathbf{M} \),

\[
H_{AF}(\theta_F - \theta_{AF}) + H_{dc}\theta_F = H_{ac}\cos(\omega t).
\]

In our measurements, the imaginary part of the transverse magnetization was always significantly smaller than its real part. Neglecting its effect on the AF dynamics in Eq. (6), we obtain

\[
Re\theta_F = \frac{(K + 1)Re\theta_{AF}}{L(\omega)},
\]

\[
Im\theta_F = \nu\omega Re\theta_F/L(\omega),
\]

where \( L(\omega) = (K + 1)^2 + (\nu\omega)^2 \). Plugging this result into Eq. (7), we obtain

\[
Re\theta_F = \frac{H_{ac}}{H_{dc} + H_{AF}[1 - (K + 1)/L(\omega)]},
\]

\[
Im\theta_F = \frac{\nu\omega H_{AF}Re\theta_F}{(H_{dc} + H_{AF})L(\omega)},
\]

describing the ac magnetization \( M_F/M \approx \theta_F \) measured in our experiment at small \( H_{ac} \).

We start our analysis of Eqs. (6) by considering simple limiting cases. At high temperatures \( T > T_N \), the effects of AF should be negligible, corresponding to \( H_{AF} \ll 0 \). In this limit, \( Re\theta_F \approx H_{ac}/H_{dc} \) and \( Im\theta_F = 0 \), as expected for a standalone \( F \). In another limit discussed in Section 11, according to Eq. (8), the dynamics of AF becomes negligible at \( \nu\omega \gg 1 \), since \( L(\omega) \rightarrow \infty \) in this limit. Equation (9) then gives \( Re\theta_F = H_{ac}/(H_{dc} + H_{AF}) \), \( Im\theta_F = 0 \), consistent with Eq. (6). In the zero-frequency limit, the viscous contribution to Eq. (6) vanishes, giving \( Re\theta_F = H_{ac}/[H_{dc} + H_{AF}(1 - 1/(K + 1))] \). This response is only weakly dependent on the value of \( K \geq 0 \), so in the calculations discussed below we use \( K = 0 \).

Figure 6 shows the calculated \( \theta_F(f) \) for the range of viscosities that result in a significant imaginary susceptibility component at the experimental frequencies \( f = 1 \text{ Hz}-7 \text{ kHz} \). The real part of \( \theta_F \) monotonically increases with decreasing frequency, with the largest variation at \( \nu \approx 1/2\pi f_0 \), where \( f_0 = 10^2 \text{ Hz} \) is the characteristic measurement frequency. Comparing with the data in Fig. 5(b), we conclude that the viscosity increases with decreasing temperature, with \( \nu \approx 10^{-2} \text{ s} \sqrt{T} \) at \( T = 240 \text{ K} \).

Because of the inhomogeneities in the studied system, associated with the variations of the local thickness, magnetic anisotropy, and the level of frustration introduced by the exchange interactions at F/AF interface, we do not expect that a single value of \( \nu \) is sufficient to quantitatively describe the behaviors at a given temperature. This is reflected by the significantly more gradual experimental variation of \( Re(M_F/M) \) with frequency. We leave the analysis of these inhomogeneous broadening effects to future studies.

According to Eqs. (6), the imaginary part of response should vanish in both the low- and the high-frequency limits, with a peak at \( \nu\omega = (K + 1) \). At \( \nu = 0.1 \text{ s} \) and \( K = 0 \), this peak is below the frequency scale in Fig. 6(b), resulting in a monotonic decrease of \( Im\theta_F \) with increasing \( f \). In contrast, at \( \nu = 10^{-3} \text{ s} \), the peak shifts close to \( f = 100 \text{ Hz} \) in the middle of the frequency range, while at \( \nu = 10^{-5} \text{ s} \), the peak shifts above 7 kHz, resulting in the monotonic increase of \( Im\theta_F \) with increasing \( f \). These results are in a good qualitative agreement with the data in Fig. 4(b) for \( T = 230 \text{ K}, 240 \text{ K}, \text{ and } 250 \text{ K} \), respectively, suggesting that the effective viscosity of AF increases by four orders of magnitude over this temperature range.

To highlight the implications of these results for the nature of the magnetization state of AF, we compare them with the prediction of the Neel-Brown theory for...
the ac susceptibility of single-domain particles that has been extensively utilized in modeling of exchange bias. According to this theory, the imaginary part of susceptibility exhibits a peak at the frequency $f_0 \approx re^{-\beta}/2\pi$, where $r$ is the attempt rate, which is close to the characteristic dynamical frequency of the magnetic nanoparticle, $\beta = U/k_BT$, and $U$ is the activation energy. The predicted rate of variation of $f_0$ with temperature is then $\frac{df_0}{dT} = \frac{f_0U}{k_BT^2}$. Based on our results, we estimate $f_0 = re^{-\beta}/2\pi \approx 10^2$ Hz and $\frac{df_0}{dT} \approx 0.2f_0n10$ at $T = 240$ K. Thus, $U/k_BT^2 \approx 0.5$ K$^{-1}$, or equivalently $\beta = 120$ at $T = 240$ K. The estimated attempt rate $r = 2\pi f_0e^{\beta} \approx 10^{55}$ s$^{-1}$ is unphysically large. This large discrepancy arises because the variation of the effective viscosity with temperature is significantly faster than would be expected from the Arrhenius activation, signifying a spin-freezing transition accompanied by an increased cooperativity of the magnetization dynamics. These results support the earlier conjecture, based on the time-domain aging measurements, that the Heisenberg domain state of the magnetization of thin AF films in F/AF bilayers is a spin glass.

V. SUMMARY

This paper presented three significant results. Our main technical result is the demonstration that the variable-temperature, variable-frequency transverse ac susceptibility susceptibility technique can provide detailed information about the anisotropy and dynamical properties of magnetic systems. In particular, we showed that the uniaxial and unidirectional anisotropies can be determined from the dependence of susceptibility on the dc field, and independently from the dependence on the ac field. Furthermore, the dependence of the real and imaginary parts of susceptibility on frequency and temperature provide information about the relaxation processes, and ultimately about the nature of the magnetization state of the system.

We utilized the transverse ac susceptibility technique to analyze the magnetic characteristics of a common exchange bias system - a thin-film CoO/Permalloy bilayer. Our first significant finding is that the exchange bias - asymmetry of the hysteresis loop with respect to the field direction - arises in the studied system not due to the small unidirectional anisotropy, but rather due to the different reversal mechanisms between the two opposite magnetization directions stabilized by a large unidirectional anisotropy. The latter likely arises due to the reversible canting (partial spin-flop) of the antiferromagnetic moments by the exchange interaction with the ferromagnet. This finding may provide a significant step towards general fundamental understanding of phenomena associated with exchange interactions at the ferromagnet/antiferromagnet interfaces, and ultimately for the ability to engineer these phenomena for applications in nanomagnetic devices.

Our third result is the observation of a spin-freezing transition in the antiferromagnet at temperatures slightly above the exchange bias blocking. This transition is signaled by a peak in the temperature dependence of the imaginary part of transverse ac susceptibility, and a significant dependence of the real part on frequency. These observations cannot be attributed to the Arrhenius-type thermal activation described by the Neel-Brown model, because this model would require an unphysically large attempt rate to accommodate the observed fast variation with temperature. This result is consistent with the conjecture, previously put forward based on the aging measurements, that the Heisenberg domain state in thin-film ferromagnet/antiferromagnet bilayers is a spin glass.

The ability to judiciously generate spin-liquid and spin-glass states in thin-film heterostructures can become extremely useful both for the fundamental research and for applications. The glass state of matter has been one of the most extensively researched subjects in condensed matter physics over the last several decades, but a universally accepted theory of this state has not yet emerged. Experimental studies of physical glasses have been hindered by the difficulty in precisely replicating the studied systems, and in identifying the physical characteristics most relevant to the glass formation. Spin glasses are straightforward to characterize and model, and the spin system can be "reset" by heating without modifying any other parameters, providing the ability of repeated measurements on identical systems. However, experimental studies have been largely limited to bulk dilute spin systems at low temperatures, making it difficult to address important questions related to the role of dimensionality of the material and of the order parameter, the effects of surfaces/interfaces, and interactions competing with the glass transition.

Spin glasses, and more generally spin systems exhibiting controllable complex collective behaviors at room temperature, can provide a suitable medium for a number of applications actively discussed by the soft condensed matter community. For instance, the stability of a multitude of configurations, determined both by the strength of the perturbations and by their history, may lend itself to efficient neuromorphic circuit implementations. We note that in contrast to ferromagnets, the density of information in spin glasses is not limited by the competition among the local exchange, dipolar, and thermal energies. Furthermore, while glassy systems are generally slow, they can also posses fast degrees of freedom that can extend beyond typical dynamical frequencies of ferromagnets, due to the relaxed angular momentum conservation conditions. Thus, such systems can share some of the advantages of AF spintronics, while providing more straightforward possibilities for the control and detection of their states.
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