Reply to “Comment on ‘Ultrafast Demagnetization Measurements Using Extreme Ultraviolet Light: Comparison of Electronic and Magnetic Contributions’ ”

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In the following, we show that the conclusions of our article titled “Ultrafast Demagnetization Measurements Using Extreme Ultraviolet Light: Comparison of Electronic and Magnetic Contributions” are correct. The Comment of Vodungbo *et al.* argues that a unique determination of the refractive index variation over time is not possible using the data set presented in our paper. Furthermore, it was suggested that the lack of uniqueness allows for the possibility of a very specific time-dependent trajectory of the refractive index in the complex plane that could give rise to a large nonmagnetic modulation of the measured asymmetry, in spite of a negligible change in the *s*-polarized reflectivity. In this Reply, we conclusively show that any nonmagnetic contribution to the measured asymmetry is indeed negligible ($< 2\%$), below the noise level of the magnetic-asymmetry measurements. First, we use a few additional measurements to unambiguously rule out the presence of any nonmagnetic contributions to the signal. Second, we show that the scenario proposed by Vodungbo *et al.* would require both exotic time and energy dependences of the refractive index near the *M* edge that are extremely unlikely (virtually impossible) in real materials. Thus, the conclusions of our original article are preserved.

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We show below that the refractive index, in response to optical pumping, does not follow the peculiar time-dependent trajectory suggested by Vodungbo *et al.* [1]. Indeed, we make use of the fact that the magnitude of the magnetic asymmetry of the transverse magneto-optical Kerr effect (T-MOKE) signal depends strongly on both the refractive index and the angle of incidence. However, for a pure magnetic signal, the dynamics should be independent of these values. As was suggested in the Comment, we first calculate the specific refractive-index contours in the complex plane that leave the *s*-polarized reflectivity constant, as observed experimentally [2,3]. We perform these calculations using both the semi-infinite approximation and also a self-consistent multilayer calculation. These *s*-polarization “isorefectivity” curves are shown in Fig. 1(a) for the case of 67.7 eV at incidence angles of 45° (green curves) and 41° (blue curves). Next, we utilize these isorefectivity curves to calculate the

magnitude of the corresponding contribution to the asymmetry due to nonmagnetic components. These plots are shown in Fig. 1(b) for the semi-infinite case and in Fig. 1(c) for both the thick- (1000-nm) and thin-film (10-nm) limits. Figures 1(b) and 1(c) predict that the asymmetries A/A_0 at 41° and 45° would strongly diverge if the refractive index *n* follows the isorefectivity trajectories of Fig. 1(a), for all film thicknesses. Thus, if we experimentally measure identical dynamics in the magnetic asymmetries A/A_0 at 45° and 41° as a function of pump fluence and of time, the proposed scenario suggested by Vodungbo *et al.* [i.e., Fig. 1(a)] can be excluded, within the accuracy of our measurements. We experimentally show below that the normalized transient asymmetry does not depend on the incident angle.

We measured the transient magnetic asymmetry as a function of laser fluence at an angle of 41° [Fig. 2(a)] and compared the extracted maximum asymmetry reduction [Fig. 2(b)] with the data in our original paper at 45° (Fig. 3 in Ref. [2]). We emphasize that we observe the same dynamic response across the full energy range of the Ni *M* edge, since our light source spans the entire extreme ultraviolet region of the spectrum. Once again, we observe a linear dependence of the maximum demagnetization

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FIG. 1. (a) Isorefectivity contours of the complex refractive index for $R^S(\delta, \beta) = R_0^S$ at two angles of incidence of 41° and 45° , for comparison with Fig. 1(a) in the Comment by Vodungbo *et al.* (b) Calculated asymmetry parameter for 41° and 45° , subject to the same isorefectivity constraint, in the semi-infinite limit. (c) Same as (b), but calculated with a conventional, self-consistent multilayer-reflectivity model, for both a thick (1000 nm) and thin film (10 nm) of Ni on 3-nm Ta. Movement of n away from point O , if restricted to the contours in (a), results in very different asymmetries for different angles of incidence. Experimentally, however, we do not observe any such dependence of the measured transient asymmetry on the incident angle (see Fig. 2).

(quenching) on pump fluence. For a purely magnetic (i.e., artifact-free) measurement, the maximum demagnetization should match the dashed red line in Fig. 2(b), since the intrinsic ultrafast magnetization dynamics must be independent of the experimental geometry (i.e., angle). As explained above, if the refractive index varies along the contour of constant s -polarization reflectivity shown in Fig. 1(a), theory predicts strongly diverging normalized transient asymmetries at incidence angles of 41° and 45° [Figs. 1(b) and 1(c)]. Therefore, any deviation of the data from the red line in Fig. 2(b) is a direct measure of the maximum possible nonmagnetic artifact introduced by the scenario proposed in the Comment. Since the deviation of

FIG. 2. (a) Demagnetization dynamics measured at a 41° angle of incidence for three different pump fluences. (b) Maximum demagnetization as a function of pump fluence. The red points are the original data from Ref. [2] measured at 45° , while the blue open circles are the new data measured at 41° . The deviation of the new data from the dashed red line, obtained from the original data via linear regression, is used to determine potential artifacts due to a pathological refractive index, which are found to be less than 2%.

the measured demagnetization in Fig. 2(b) at both 41° and 45° is less than 2%, we conclude that any potential non-magnetic modulation of the measured asymmetry due to a pathological time dependence of n is still less than the noise in our magnetic-asymmetry measurements ($\leq 2\%$). Thus, our data indicate that the refractive index n must stay at the value indicated by point O in Fig. 1 and does not follow the contour suggested by Vodungbo *et al.* As a result, the final conclusion drawn from our original paper is unchanged: The observed reduction of the magnetic asymmetry in response to optical pumping is purely magnetic in origin.

In addition, we note that the isorefectivity constraint for n proposed by Vodungbo *et al.* not only leads to a strong

dependence of the measured asymmetry on incident angle but also to a strong dependence of the p -polarized reflectivity on angle, which we do not observe. Finally, the specific time-varying trajectories of the complex refractive index required to minimize changes of the s -polarized reflectivity would also depend strongly on energy near the M edge, given the strong dependence of n on energy at an absorption edge. Thus, for the scenario proposed by Vodungbo *et al.* to be true, the refractive index must not only follow a particular time-dependent trajectory in the complex plane but an extremely pathological, energy-dependent trajectory such that the s -polarized reflectivity change is independent of both time and energy. Not only are we unaware of any material with such bizarre properties, but the existence of any such material seems unlikely in the extreme.

Finally, we address the minor criticisms presented in the Comment.

- (i) We did not indicate which line of Eq. (3) was used for the asymmetry calculations.

Answer: We used line 1. However, the quantitative differences in the results obtained by use of either line 1 or 2 are small.

- (ii) The third line of Eq. (3) cannot be derived by a proper expansion of line 2.

Answer: It is indeed the case that we utilized the additional unstated approximation

$$(1 - 2\cos^2\theta)^2 \gg |1 + 8\cos^2\theta - 12\cos^4\theta|\delta, \quad (1)$$

which allowed us to only retain terms to second order in δ , β in the denominator, where $n = 1 - \delta + i\beta$ ($\delta, \beta > 0$). If we use $\delta = 0.01066$ and $\beta = 0.1003$, as obtained from tabulated data at 67.7 eV [4], the approximation is valid over a wide range of angles, excluding a narrow span of $\pm 6^\circ$ centered at 45° . In addition, we wish to stress that the purpose of line 3 in Eq. (3) was to simply demonstrate the resonant nature of the T-MOKE signal at the M edge. No approximations were used in the subsequent calculations, nor are they used for the extended analysis presented in this Reply.

- (iii) Equation (1) (of Ref. [2]) is only valid for a semi-infinite magnetic plane.

Answer: We have repeated the calculations by use of a self-consistent, Fresnel-based multilayer-reflectivity model that includes magneto-optic effects [5]. The calculations were performed for both 10- and 1000-nm Ni thicknesses, as shown in Fig. 1 of this Reply. While it is the case that the quantitative dependence of the magnetic asymmetry on the refractive index is affected by finite thickness for our measured samples, we also find that the qualitative dependence is, as expected, unchanged: The dependence of the magnetic asymmetry on the refractive-index change is a strong function of

incident angle, making it a very straightforward exercise to exclude the remote possibility of pathological time-varying trajectories of the refractive index in the complex plane, as discussed above.

- (iv) We present data for s -polarization reflectivity obtained at 66.2 eV, while our T-MOKE asymmetry data is for 67.7 eV.

Answer: By use of broadband extreme ultraviolet light, we have the unique and powerful ability to measure the reflectivity across the entire M edge as a function of time. Thus, we collect and analyze data for all photon energies [6–8] and carefully compare the dynamic response across the entire M edge. We plot the data with the best signal-to-noise ratio for a given photon energy, having first carefully determined that we measure here the same normalized changes in reflectivity and asymmetry at all photon energies near the M edge.

- (v) The magneto-optical constant ε_{xy} , which is needed for the calculation of the asymmetry, is not given.

Answer: We used the value $\varepsilon_{xy} = -0.007 - i0.0066$. This value was extracted from data in Ref. [25] in our original paper, again by use of the tabulated refractive index at 67.7 eV. However, our results or general conclusions are not strongly dependent on the exact value of the magneto-optic parameter for this system.

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