Terahertz magneto-optical response in ferromagnetic Fe-Co-Al alloys

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We study the magneto-optical properties of Fe-Co-Al ordered alloys in the terahertz range of frequencies. Using the standard Kubo-based approach to compute intrinsic part of the $\sigma_{xy}(\omega)$ we find a strong dependence of σ_{xy} on ω in the terahertz range. For example, we find that below 10 THz Co₃Al has nearly constant σ_{xy} and that above 10 THz it is reduced by about 50 times. Furthermore, we find a strong dependence of σ_{xy} on the chemical composition. For example, we find that the addition of Al to Fe changes the sign of σ_{xy} , while the addition of Co to Fe leads to a nonmonotonic dependence of σ_{xy} on Co concentration.

I. INTRODUCTION

A magnet with a cubic crystal structure and magnetization pointing along the z-axis has a non-zero offdiagonal component of the optical conductivity, $\sigma_{xy}(\omega)$. This contribution to the conductivity occurs because magnetic order in such a system breaks time-reversal symmetry. The breaking of the time-reversal symmetry is propagated to the electronic degrees of freedom by the spin-orbit interaction. Therefore, the magnitude of $\sigma_{xy}(\omega)$ is dictated by the spin-orbit interaction strength.¹ Presence of off-diagonal conductivity σ_{xy} , and thus indirectly the presence of magnetic order, can be detected optically by comparing the polarization of light incident to, and reflected from, the surface of a magnet. This occurs for example in the so-called magneto-optical Kerr effect, MOKE.² Opposite is also true. If one changes the magnetization direction along the z-axis,

$$M_z \to -M_z,$$

this will result in,

$$\sigma_{xy} \to -\sigma_{xy}$$

Therefore, one can use the direction of magnetization to control the way in which light reflects from a surface of a magnet. This effect has been used in memory storage devices^{3,4} and in the creation of tunable photonic materials.^{5,6}

At zero frequency, the static conductivity $\sigma_{xy}(\omega = 0)$ produces the anomalous Hall effect (AHE). In materials with very few impurities, so that the diagonal conductivity σ_{xx} is above $10^6 \ (\Omega \ cm)^{-1}$, the dominant contribution to AHE originates from the scattering of electrons from the impurities. On the other hand, somewhat paradoxically, in materials with a moderate amount of impurities, with diagonal conductivity around $10^4-10^6 \ (\Omega \ cm)^{-1}$, the dominant contribution to AHE is intrinsic, independent of the number of impurities.¹ The intrinsic contribution to AHE is given by the integral of the Berry curvature over the occupied states.⁷ However, in the dynamic case, the $\sigma_{xy}(\omega \neq 0)$ can no longer be written as a sum over the Berry curvature, instead it needs to be computed from the Kubo-like sum over empty states, as in Refs. 8–11. Nevertheless, we expect that for small enough ω the intrinsic contribution still dominates $\sigma_{xy}(\omega)$, which is consistent with findings in Ref. 12.

There are many calculations of static intrinsic AHC in the literature. For example, Yao¹³ calculated the AHC of ferromagnetic bcc Fe, and the calculated σ_{xy} at zero frequency is 751 (Ω cm)⁻¹. Wang¹⁴ found that at zero frequency, AHC (σ_{xy}) in bcc Fe, fcc Ni, and hcp Co is 753, -2203, and 477 (Ω cm)⁻¹, respectively. Bianco¹⁵ calculated the AHC in Fe₃Co, and the σ_{xy} value at zero frequency is 452 (Ω cm)⁻¹. Huang¹⁶ calculated the AHC of the Heusler compound such as Co₂FeAl and found it to be 39 (Ω cm)⁻¹.

While σ_{xy} originates from magnetism, the proportionality coefficient between σ_{xy} and the magnetic moment is difficult to predict without performing an explicit firstprinciples calculation. As discussed in early work on frequency dependence of σ_{xy} from Ref. 9, as well as in later Refs. 13–15, and 17, different parts of the reciprocal space can have either positive or negative contribution to σ_{xy} , as sign will in general depend on position of the Fermi level relative to the subtle spin-orbit induced band gaps in the band-structure. Therefore, we might expect that, generally σ_{xy} will be a very sensitive function of the electronic band structure. As a consequence, we expect a rich dependence of dynamic $\sigma_{xy}(\omega)$ in the low-frequency range. In particular, we expect that there will be a strong frequency dependence of $\sigma_{xy}(\omega)$ when $\hbar\omega$ is close to the energy of the spin-orbit split bands in the band structure. The energy of the spin-orbit split bands is on the order of tens of meV in ferromagnetic metals such as Fe, Co, or Ni. These energies lie in the range of frequencies ~ 10 THz, within the so-called *terahertz-gap*: the range of frequencies in the electromagnetic spectrum that are in between the microwave radio frequencies and optical frequencies.

Moreover, we quite generally expect that σ_{xy} will be very sensitive on alloying, as even subtle changes in the electronic band structure will change the position of spinorbit split bands relative to the Fermi level. In this study, we use first-principles techniques to calculate the $\sigma_{xy}(\omega)$ in the THz regime for a specific ternary metallic alloy system, Fe-Co-Al. We decided to focus on this alloy in particular, as it is known that the addition of Co to Fe leads to strong variations of spin-polarized density of states, as studied, for example, in Ref. 18. Furthermore, even small addition of Al to Fe–Co can lead to large changes in the measured spin-dependent physical properties, as shown in Refs. 19–21. Our calculations show that alloying is indeed a feasible way to change the THz optical response of ferromagnetic metals in the Fe-Co-Al ternary system.

The early work from Ref. 9 focuses on the frequency dependence of σ_{xy} in Fe, Co, and Ni for frequencies ω above 0.1 eV. Most other studies of frequency-dependent σ_{xy} are carried out in the optical regime, at even higher energies. For example, Ref. 22 computed σ_{xy} for Fe₃Co and FeCo₃ in the 0.5–5.2 eV energy range. Next, Ref. 23 calculated the optical response of $\operatorname{Fe}_{4-x}\operatorname{Co}_x$ (with x = 1-3) up to the 13 eV range, while Ref. 24 and 25 reported σ_{xy} for FeAl in the 0.5–6 eV range. Recently, some studies have focused on the off-diagonal optical response in the terahertz (THz) regime. Seifert²⁶ studied the off-diagonal optical response in $DyCo_5$, $Co_{0.32}Fe_{0.68}$ and $Gd_{0.27}Fe_{0.73}$, and they measured somewhat stronger dependence of σ_{xy} on ω in the range of frequencies below 10 THz, than above 10 THz. Matsuda¹² studied the off-diagonal optical response in Weyl antiferromagnet Mn₃Sn at very low energy (10 meV) and they find weak dependence of σ_{xy} on frequency. On the other hand, calculated and measured σ_{xy} in the THz regime is very frequency dependent in SrRuO₃, as shown in Refs. 27–29.

We organize the paper as follows. In Sec. II we show the calculation methods. In Sec. III we present and analyze our results. We conclude in Sec. IV.

II. METHODS

We use the Quantum Espresso package³⁰ to calculate the electronic structure of ordered alloys of Fe, Co, and Al. We use the Generalized Gradient Approximation (GGA) of Perdew, Burke, and Ernzerhof (PBE)³¹ along with the optimized norm-conserving Vanderbilt (ONCV) pseudopotentials which include spinorbit interaction.^{32–34} We choose 120 Ha kinetic-energy cutoff for the plane-wave expansion of the valence wave functions. A $16 \times 16 \times 16$ Monkhorst-Pack grid and a smearing³⁵ of 0.01 Ry are used to sample the electron's Brillouin zone. We computed $\sigma_{xy}(\omega)$ using the standard Kubo formula,³⁶

$$\sigma_{\alpha\beta}(\omega) = \frac{ie^2\hbar}{VN_k} \lim_{\delta \to 0} \sum_{\mathbf{k}} \sum_{nm} \frac{f_{m\mathbf{k}} - f_{n\mathbf{k}}}{\varepsilon_{m\mathbf{k}} - \varepsilon_{n\mathbf{k}}} \frac{\langle \phi_{n\mathbf{k}} | v_\alpha | \phi_{m\mathbf{k}} \rangle \langle \phi_{m\mathbf{k}} | v_\beta | \phi_{n\mathbf{k}} \rangle}{\varepsilon_{m\mathbf{k}} - \varepsilon_{n\mathbf{k}} - \hbar\omega - i\delta/2} \tag{1}$$

that was already used successfully in Refs. 8–11. Here, α and β denote Cartesian directions. V is the cell volume, N_k is the number of k-points, and $f_{n\mathbf{k}}$ is the Fermi-Dirac distribution function. ω is the optical frequency. We use the Wannier interpolation^{37,38} in evaluating Eq. 1, as these calculations require a very dense sampling of the Brillouin zone. We tested the convergence of σ_{xy} with the choice of the k mesh. We use adaptive k-mesh refinement to accelerate convergence¹³ by adding a 5×5×5 fine mesh around regions with a large contribution to $\sigma_{xy}(\omega)$. For pure metals (Fe and Co), a 250×250×250 primary k-mesh is enough to achieve the convergence of σ_{xy} . For calculations with two atoms per unit cell and four atoms per unit cell, a 200×200×200 and 150×150×150 k-mesh, respectively, was enough to achieve convergence.

In this work we mostly stay within the intrinsic limit $\delta \to 0$ of the σ_{xy} for small ω , as given by Eq. 1. For future work we leave the role of finite electron lifetime, random disorder, or temperature effects on $\sigma_{xy}(\omega)$ in the THz regime. As discussed in Sec. IV we attempt to approximately model disorder within the approach of Ref. 39.

To validate the reliability of our calculation approach, we compare our calculation results with previous calculations. Our calculated σ_{xy} of Fe and Co at zero frequency are 758 and 471 (Ω cm)⁻¹, respectively, which agrees very well with the previous calculation result of 753 ($\Omega \text{ cm}$)⁻¹ in Fe and 477 ($\Omega \text{ cm}$)⁻¹ in Co.¹⁴ Furthermore, the calculated σ_{xy} of FeCo and Fe₃Co at zero frequency is 226 and 416 ($\Omega \text{ cm}$)⁻¹, respectively. The measured σ_{xy} of Fe_{0.68}Co_{0.32} at zero frequency is about 350 ($\Omega \text{ cm}$)⁻¹²⁶, which lies between the theoretical values of FeCo and Fe₃Co. Furthermore, we calculated the Kerr angle of Fe in the range of 0–1.2 eV by using our calculated σ_{xy} values and experimental σ_{xx} values from Ref. 40 and 41. Our calculated Kerr angle matches very well with the results in Ref. 42.

In all of our calculations, we assumed that the magnetization points along the [001] direction. We did not explore what happens to the off-diagonal conductivity when the magnetization points in any other crystallographic direction, such as [011] or [111]. We decided to restrict our calculations to those with the magnetization axis pointing along the [001] direction, as our goal here is to compare σ_{xy} only as a function of the chemical composition of the alloy and frequency. Furthermore, for consistency, in each calculation we chose the same sign of magnetization along the [001] direction.

Now we present our results for ordered Fe-Co-Al alloys. We start by discussing the computed lattice constants and crystal structure of these alloys. Our results are shown in Table I. Most of the ordered alloys we studied order in the bcc-derived structures B_2 and $D0_3$.^{43–45} The \mathbf{B}_2 structure is a bcc-derived structure with two atoms in the primitive unit cell. This structure is therefore present in alloys with the ratio of 1–1 of two elements. On the other hand, $D0_3$ structure contains four atoms in the primitive unit cell, so it is present in ordered binary alloys with the 1–3 ratio of constituent elements, or the 1–1–2 ratio in the case of ternary alloys. Among the ordered alloys we studied, the only ones that are not in the bcc-derived structure are Al, FeAl₃, CoAl₃, Co, and Co_3Al . However, three of these (Al, FeAl₃, and CoAl₃) are nonmagnetic, regardless of their crystal structure, so their σ_{xy} is identically zero. Therefore, we don't discuss these cases in more detail. Next, in the case of Co, we explicitly showed that $\sigma_{xy}(\omega)$ is very similar in bcc and hcp structures. Therefore, for a more consistent comparison with other members of the Fe-Co-Al family of compounds, we will show results for pure Co in its bcc structure. Finally, we expect that the remaining exception, Co-rich Co₃Al, will also have $\sigma_{xy}(\omega)$ that doesn't strongly depend on the structure, so that we are justified in studying Co_3Al in the bcc-derived structure (and not in the lowest energy structure, the fcc-derived $L1_2$).

We find that the computed lattice constants are close to the values experimentally measured. Small deviations, on the order of 1% are due to the approximations in our exchange-correlation functional, as well as thermal expansion, as the experimental data in Table I are taken at room temperature.

TABLE I. Calculated magnetic moment and lattice constant of ordered Fe-Co-Al alloys. The magnetic moment is on a per atom basis, and this includes either nominally magnetic atom (Fe, Co), or non-magnetic atom (Al).

		M	a^{calc}	a^{\exp}
		$(\mu_{\rm B}/{\rm atom})$	(Å)	(Å)
Fe	bcc	2.26	2.84	2.84
Fe_3Co	$D0_3$	2.36	5.71	5.66
FeCo	B_2	2.30	2.85	2.85
$FeCo_3$	$D0_3$	2.02	5.66	5.66
Co	bcc	1.80	2.82	2.82
Ca	hon	1.69	a = 2.50	a = 2.50
CO	ncp	1.02	c = 4.04	c = 4.09
Fe ₃ Al	$D0_3$	1.55	5.76	5.79
FeAl	B_2	0.36	2.88	2.91
FeAl ₃	$D\theta_3$		5.98	
Co ₃ Al	$D\theta_3$	1.05	5.69	
CoAl	B_2		2.86	2.86
CoAl ₃	$D\theta_3$		6.01	
Fe ₂ CoAl	$D0_3$	1.54	5.76	5.73
FeCo ₂ Al	$D0_3$	1.25	5.71	5.73

A. Fe and Co

Figure 1 shows calculated $\sigma_{xy}(\omega)$ of pure Fe and Co metal in the range of energies from $\hbar\omega~\sim~0\text{--}0.1$ eV. This corresponds to the range of frequencies $\omega/(2\pi) \sim 0$ -25 THz. As expected, we find a strong variation of $\sigma_{xy}(\omega)$ as a function of frequency, in both Fe and Co. These variations are the strongest around 10–15 THz. We attribute these modifications to the fact that the characteristic spin-orbit gaps in these metals occur in the same range of energies. For Fe, we find that the minimal value of σ_{xy} , in the studied frequency range, is 634 $(\Omega \text{ cm})^{-1}$ at 14.5 THz, while the maximal value is 1.5 times larger, 950 $(\Omega \text{ cm})^{-1}$ at a nearby frequency of 16 THz. In the case of Co, the minimal value is 233 $(\Omega \text{ cm})^{-1}$ at 17 THz, and maximal value is 2.5 times larger, 582 $(\Omega \text{ cm})^{-1}$ at 10 THz. We compared $\sigma_{xy}(\omega)$ in bcc and hcp structure of Co and we found qualitatively similar responses, as shown in Fig. 1. Therefore, at least in the case of Co, the crystalline structure doesn't have a strong effect on $\sigma_{xy}(\omega).$

B. Fe-Co alloys

After analyzing σ_{xy} in pure Fe and Co, we now turn to the ordered Fe-Co alloys. We considered three ordered alloys of Fe and Co, these are Fe₃Co, FeCo, and FeCo₃. The ordered crystalline structures of these alloys are bccderived D0₃, B₂ and D0₃, respectively. Our results for Fe-Co alloys are shown in Fig. 2. Starting from pure Fe, we find that the addition of Co at first significantly reduces the value of σ_{xy} . In particular, σ_{xy} in Fe₃Co is on average about 2 times lower than that of pure Fe.

Calculated magnetic moments per atom are also given in Tab. I. Most of these ordered alloys are magnetic in our calculations, with the exception of Al-rich compounds, such as FeAl₃, CoAl₃ and CoAl. These findings are in agreement with previous studies.^{46,47}



FIG. 1. Calculated real part of σ_{xy} for bcc Fe, bcc Co, and hcp Co as a function of ω .



FIG. 2. Calculated real part of σ_{xy} for bcc Fe, D0₃ Fe₃Co, B₂ FeCo, D0₃ FeCo₃ and bcc Co as a function of ω .

Furthermore, the spectral features of alloy Fe₃Co are distinct from that of pure Fe. While both Fe and Co have a nearly constant σ_{xy} up to 10 THz, we find that Fe₃Co shows a strong dependence on frequency starting already around 3 THz. This finding of a large sensitivity of σ_{xy} to the chemical composition is in agreement with our expectation that σ_{xy} is very sensitive to details of the band structure. Adding even more Co, as in FeCo, we find that the value of σ_{xy} is reduced even further. In particular, σ_{xy} is about 4 times smaller in FeCo than in pure Fe. Moreover, the spectral features of FeCo are surprisingly constant in the entire range of frequencies we studied. The further addition of Co increases the value of σ_{xy} . For example, in the case of FeCo₃, σ_{xy} is similar in magnitude to that of pure Co. The spectral features of FeCo₃ are the strongest among the Fe-Co alloys we studied.

The nonmonotonic dependence of σ_{xy} on Co concentration is reminiscent of a nonmonotonic dependence of



FIG. 3. Calculated real part of σ_{xy} for bcc Fe, D0₃ Fe₃Al, B₂ Fe_{0.50}Al_{0.50} and bcc Al as a function of ω .

magnetic damping observed in Fe-Co alloys.¹⁸ Although magnetic damping is not directly related to σ_{xy} , we expect that both can be related to changes in the nature of the electronic band structure near the Fermi level in Fe-Co alloys.

We note that variation in σ_{xy} on Co concentration can't be rationalized with variation of the magnetic moment, as we find that all members of the Fe-Co family of alloys have nearly constant magnetic moment per atom, on the order of ~ $2\mu_{\rm B}$ (see Table I). Therefore, even though σ_{xy} can be used as a magnetic order signature, one is not necessarily proportional to the other. As discussed earlier, the role of the magnetic order here is only to break the time-reversal symmetry, while the magnitude and spectral properties of σ_{xy} are driven by the spin-orbit interaction of electron bands close to the Fermi level.

Comparing the band structure of all Fe-Co ordered alloys we studied, we find that the overall band structure has not changed much, but only the position of the Fermi level relative to the rigid band structure is increasing with the addition of Co. Therefore, we attribute the changes in the σ_{xy} of Fe-Co alloys to changes in the relative position of the Fermi level, and not to changes in the band structure itself.

C. Fe-Al alloys

Next, we discuss the calculated σ_{xy} in Fe-Al alloys. Our results are shown in Fig. 3. Since Al is non-magnetic, the σ_{xy} response is, by symmetry, zero at all frequencies. When Al is introduced, the σ_{xy} response of Fe₃Al lies between that of pure Fe and Al, as expected. However, unexpectedly, when the Al concentration reaches 50%, σ_{xy} response of FeAl changes sign in the entire frequency range from 0 to 20 THz. We ensured that, in all of these cases, the Fe and Fe-Al alloys have magnetization point-



FIG. 4. Calculated real part of σ_{xy} for bcc Co, D0₃ Co₃Al and bcc Al as a function of ω .

ing in the same direction. A similar result is observed in Co/Pd multi-layers in Ref. 48, where the sign of σ_{xy} changes depending on the relative concentration of Co to Pd.

D. Co-Al alloys

The next binary alloys we discuss are Co-Al alloys. Results for these alloys are shown in Fig. 4. Our calculations find that CoAl and CoAl₃ are not magnetic while the Co-rich compound Co₃Al is magnetic. At low frequencies, below 10 THz, we find that σ_{xy} for Co₃Al is quite large, and somewhat constant, with the value of ~ 500 (Ω cm)⁻¹. However, above 10 THz, σ_{xy} is reduced 50-fold to only ~ 10 (Ω cm)⁻¹.

E. Fe-Co-Al ternary alloys

So far, we discussed the σ_{xy} response of binary alloys. In what follows, we consider several Fe-Co-Al ternary alloys. We kept the Al concentration at 25%, and varied the relative concentration of Fe and Co. The results for these alloys are shown in Fig. 5. The behavior of Fe_2CoAl is qualitatively similar to that of Fe_3Al , so the replacement of Fe with Co did not change qualitatively $\sigma_{xy}(\omega)$. Quantitatively, we find that σ_{xy} is approximately ~ 1.5 times lower in Fe₂CoAl relative to Fe₃Al. However, with an even higher concentration of Co, as in $FeCo_2Al$, we find $\sigma_{xy}(\omega)$ which is qualitatively and quantitatively different from other members of the Fe-Co-Al family of compounds. In particular, we find an unusually small σ_{xy} in FeCo₂Al, around 80 (Ω cm)⁻¹, that is almost insensitive to the frequency ω in the entire range between 0 and 25 THz.



FIG. 5. Calculated real part of σ_{xy} for D0₃ Fe₃Al, D0₃ Fe₂CoAl, D0₃ FeCo₂Al and D0₃ Co₃Al as a function of ω .

F. Optical conductivity and magnetic moment

As we discussed earlier, σ_{xy} is often taken as an optical signature of magnetic order. Therefore, it is natural to ask whether a material with a large magnetization will also have a large σ_{xy} . Figure 6 shows the relationship between σ_{xy} and the magnetic moment per atom for all of the compounds we studied. For each compound we show on the vertical scale of Fig. 6 the range of maximal and minimal values of the calculated σ_{xy} in the range of frequencies from 0 to 25 THz. As can be seen from the figure, we find that in the case of Fe-Co alloys, the magnetic moments per atom are nearly the same, on the order of ~ 2 $\mu_{\rm B}$, but the σ_{xy} ranges from 150, all the way to 950 $(\Omega \text{ cm})^{-1}$. The same is true for the other compounds we studied. The most drastic example is FeAl in which σ_{xy} even changes sign relative to that of Fe. Therefore we can conclude that materials with larger magnetic moment do not necessarily have larger σ_{xy} . This is not too surprising, as the large magnetization arises from the large difference in population of dominantly spin-up and spindown bands. However, a large σ_{xy} in the low-frequency regime relies on detailed information about spin orbit split bands near the Fermi level.

G. Remaining components of the σ tensor

So far we have focused on the frequency dependence of the real part of σ_{xy} in Fe-Co-Al ternary system. However, there are other changes to the conductivity tensor induced by the magnetic order. These are the imaginary part of the off-diagonal component σ_{xy} , as well as the difference between the diagonal components of the conductivity tensor along and perpendicular to the direction of the magnetic moment, $\sigma_{zz} - \sigma_{xx}$.

We first briefly discuss the imaginary part of σ_{xy} which

contributes to the magneto-optical Kerr effect, MOKE, in addition to the real part of σ_{xy} . The imaginary part of σ_{xy} is shown in the supplement for all the compounds we studied. As expected from the Kramers-Kronig relationship, the imaginary part of σ_{xy} is also strongly dependent on both chemical composition and frequency. For example, we find that the step-like spectral feature we found for the real part of σ_{xy} in Co₃Al is accompanied with a single sharp feature in the imaginary part of σ_{xy} , at nearly the same frequency (~ 10 THz), as one might expect. Similarly, sharp features in the imaginary part of σ_{xy} we find in the case of Co and Fe₃Al. The spectral features of the imaginary part of σ_{xy} are much more complex for the other compounds we studied. This is particularly true for alloys containing Fe magnetic atoms.

For completeness, we now analyze the remaining component of the conductivity tensor that depends on the presence of the magnetic order. By symmetry, magnetization along the z axis introduces a spin-orbit driven difference between the σ_{zz} (along the magnetization axis) and $\sigma_{xx} = \sigma_{yy}$ (perpendicular to the magnetization axis). While the difference $\sigma_{zz} - \sigma_{xx}$ does not contribute to the magneto-optical Kerr effect (MOKE) it does contribute to the second order change in the birefringence (Voigt effect). While σ_{xy} is zero without magnetic order, the diagonal components $\sigma_{xx} = \sigma_{yy}$ and σ_{zz} are not. Therefore, here we don't focus on these diagonal components individually, but instead we focus on their difference, $\sigma_{zz} - \sigma_{xx}$. The calculated values of $\sigma_{zz} - \sigma_{xx}$ for all compounds we studied can be found in the supplement. Again, we find strong variations of $\sigma_{zz} - \sigma_{xx}$ both as a function of frequency and as a function of chemical composition. In the case of the Fe-Co family of compounds, we find that $\sigma_{zz} - \sigma_{xx}$ in Fe and FeCo have a sharp peak around 15 THz, and are nearly zero below 14 THz. Co shows two sharp features, with opposite signs, one at around 9 and another around 20 THz. The addition of 25% of Al to Fe strongly changes the value of $\sigma_{zz} - \sigma_{xx}$. While Fe has a sharp feature around 15 THz, Fe₃Al has a sharp feature around 3 THz. The addition of even more Al, as in FeAl, introduces two sharp features, at around 10 and 15 THz. The addition of Al to Co also introduces significant qualitative and quantitative changes to $\sigma_{zz} - \sigma_{xx}$.

IV. DISCUSSION AND CONCLUSION

We find a strong dependence of $\sigma_{xy}(\omega)$ in the terahertz frequency range for Fe-Co-Al ordered alloys. For example, in the case of Co₃Al we find a nearly 50-fold reduction in $\sigma_{xy}(\omega)$ above 10 THz as compared to $\sigma_{xy}(\omega)$ below 10 THz. On the other hand, in the case of FeCo₂Al we find a nearly constant $\sigma_{xy}(\omega)$ in the entire range from 0 to 25 THz. Furthermore, we also find a strong dependence of $\sigma_{xy}(\omega)$ on the chemical composition. For example, the addition of Al to Fe can change the sign of $\sigma_{xy}(\omega)$, so that $\sigma_{xy}(\omega)$ is positive in Fe and Fe₃Al but



FIG. 6. The summary of σ_{xy} in the range of 0.0–0.1 eV and magnetic moment per atom of each metal and alloy. The bar shows the maximum and minimum of the σ_{xy} in the range of 0.0–0.1 eV.

negative in the case of FeAl. Similarly, the addition of Co to Fe produces a nonmonotonic dependence of $\sigma_{xy}(\omega)$ on Co concentration. As an example, $\sigma_{xy}(\omega)$ in FeCo is about 4 times smaller than in Fe and 3 times smaller than that in Co. We attribute both of these strong variations, with frequency and composition, to the subtle changes in the electronic structure induced by the presence of the spin-orbit interaction. However, unlike in the case of AHE, the analysis of the origin of $\sigma_{xy}(\omega)$ is additionally complicated by the fact that the Kubo expression for $\sigma_{xy}(\omega)$ involves a transition between empty and occupied states, while in the case of AHE $\sigma_{xy}(\omega = 0)$ can be written as integral of the Berry curvature over occupied set of bands. Therefore, we leave a more detailed analysis of the origin of these strong variations of $\sigma_{xy}(\omega)$ to future studies. Here we only briefly comment on the relationship between $\sigma_{xy}(\omega)$ and the changes in the electron bands caused by the spin-orbit interaction. In the supplement we report the joint density of states (JDOS) with and without inclusion of the spin-orbit interaction. We find that whenever $\sigma_{xy}(\omega)$ experiences a large change as a function of ω , there is a corresponding spike in the JDOS due to the inclusion of the spin-orbit interaction. Therefore, the gaps in the electron spectrum induced by spin-orbit interaction are well correlated to $\sigma_{xy}(\omega)$, as expected.

Our calculations are performed in the limit of infinite electron lifetime. While this is justified for materials with moderate amount of disorder, with diagonal conductivity around 10^4-10^6 (Ω cm)⁻¹, we leave for future work discussion of role of disorder on $\sigma_{xy}(\omega)$ in the terahertz range for materials that are not within the moderate range of disorder. We expect that at low enough frequencies the phenomenology of $\sigma_{xy}(\omega)$ will be the same as that of $\sigma_{xy}(\omega = 0)$, so that with a moderate amount of disorder the dominant contribution to $\sigma_{xy}(\omega)$ is intrinsic, but with less disorder scattering from impurities start to dominate.¹ We show in supplement $\sigma_{xy}(\omega)$ with approximately incorporated effect of the finite carrier lifetime. While the finite carrier lifetime approximation washes out some of the spectral features in $\sigma_{xy}(\omega)$, we still find that many qualitative characteristics remain, such as the change in sign of σ_{xy} near 20 THz, or the sharp decline in σ_{xy} in Co₃Al around 10 THz, or non-monotonic dependence of σ_{xy} of Co concentration in Fe-Co alloys.

Our findings indicate that alloys such as Fe-Co-Al would be of interest in the creation of magnetic optical

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metamaterials in which the direction of magnetic moment and chemical composition are used to control its interaction with light.

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Supplemental Information for "Terahertz magneto-optical response in ferromagnetic Fe-Co-Al alloys"

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FIG. 1. Calculated imaginary part of σ_{xy} for Fe-Co-Al metals and alloys as a function of frequency ω . All conventions are the same as in the main text.

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FIG. 2. Calculated real part of σ_{zz} - σ_{xx} for Fe-Co-Al metals and alloys as a function of frequency ω . All conventions are the same as in the main text.



FIG. 3. Calculated real part of σ_{xy} for Fe-Co-Al metals and alloys with a simplified model that incorporates electron lifetime. The range of frequencies in this plot is larger than that in other parts of the manuscript. Frequencies go from 0 to 125 THz. We use model from Ref. 1 to approximately incorporate effect of electron lifetime into our calculation, as it is well known² that simply using $\omega + i\delta$ for finite δ leads to the wrong dependence of conductivity in metals at low frequencies. Following the model from Ref. 1 we replace $\lim_{\delta \to 0} (\ldots \omega + i\delta/2)^{-1}$ in our Kubo formula in the main text with $\sqrt{\omega(\omega + i\delta)}$ and we use a finite value, $\delta = 0.2$ eV that is close to the average inverse electron lifetime in Fe-Co-Al alloys.



FIG. 4. Calculated joint density of states of Fe-Co metals and alloys as a function of frequency ω . Grey solid lines are the joint density of states with spin-orbit coupling and grey dashed lines are the joint density of states without spin-orbit coupling.



FIG. 5. Calculated joint density of states of Fe-Al metals and alloys as a function of frequency ω . Grey solid lines are the joint density of states with spin-orbit coupling and grey dashed lines are the joint density of states without spin-orbit coupling.



FIG. 6. Calculated joint density of states of Co-Al metals and alloys as a function of frequency ω . Grey solid lines are the joint density of states with spin-orbit coupling and grey dashed lines are the joint density of states without spin-orbit coupling.



FIG. 7. Calculated joint density of states of Fe-Co-Al alloys as a function of frequency ω . Grey solid lines are the joint density of states with spin-orbit coupling and grey dashed lines are the joint density of states without spin-orbit coupling.