TUT Magnetic Materials Laboratory Seminar

豊橋技科大磁性研究室セミナー02/07/25

Recent Topics in Magneto-Optics Linear and nonlinear magneto-optical effects in Fe/Au and Co/Ru superlattices

磁気光学研究の最近の¥話題 Fe/Au, Co/Ru人工格子の線形および非線形磁気光学効果

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#### Introduction はじめに

- Two-types of superlattices
  - TM/Cu, Ag, Au →immiscible; abrupt interface (非固溶;急峻な界面)
  - TM/Pt, Pd→ miscible, gradual interface
     (固溶系:界面合金化)
- Superlattices and characteristic length (人工格子・多層膜と特性長)
  - L\*~ $\lambda$ '(light wavelength光の波長): MO enhancement
  - L\*~ds(roughness界面の荒さ)→相互拡散、合金化
  - L\*~ λ D(de Broglie wavelenth)→Quantum confinement 量子閉じこめ
  - L\*~a (atomic size)→band modificationバンドの改変

Linear magneto-optical effect in Fe/Cu compositionally modulated multilayers Fe/Cu組成変調多層膜の磁気光学効果

- Layer thickness ~ wavelength [波長( $\lambda'$ )~層厚(d)]
  - Plasma enhancement (プラズマ端でのエンハンス効果)
  - Roughly explained by effective permeability (実効誘電 率)
  - Multiple reflection and interference(多重反射·干涉効果)
  - Unaccountable for d<a few nm (変調周期が数nm以下に なると説明できなくなる)

mutual diffusion and alloy formation at the interface



Experimental magnetooptical and reflectivity spectra in Fe/Cu multilayer with different layer thickness 種々の層厚をもったFe/Cu 組成変調多層膜の磁気光学 スペクトルおよび反射スペ クトル(実験値)

#### Virtual optical constant method(仮想光学定数の方





Calculated magnetooptical and reflectivity spectra in Fe/Cu multilayer with different layer thickness 種々の層厚をもったFe/Cu組成 変調多層膜の磁気光学スペク トルおよび反射スペクトル (計算値)

Modulation period dependence of Kerr rotation in Fe/Cu multilayers (▲△experiments,solid and broken line: calculation) Fe/Cu組成変調多層膜のカー回転角の変調周期依存性。



Magneto-optical Effect in Au/Fe/Au trilayer Au/Fe/Au三層超薄膜の磁気光学効果

- Layer thickness ~ de Broglie wavelength of electrons

   層厚が電子のドブロイ波長と同程度になった場合
- New magneto-optical transition in epitaxially grown Au(cap)/Fe(ultra thin layer)/Au(buffer)/MgO(substrate) trilayer structure MBE法でMgO基板上にエピタキシャル成長したAu(100)薄膜の 上にFe超薄膜を作製し、その上に保護層としてAuの薄い キャップ層をかぶせた三層膜における新しい光学遷移
- At first the optical structure was assigned to 2D-band. Afterward it was re-explained in terms of quantum confinement of electrons in Fe-layer 当初:2Dのバンドによると同定→その後、Fe層内での電子の 量子閉じこめによるとして説明された。



#### Kerr rotation spectra in Au/Fe/Au ultra thin films Au/Fe/Au超薄膜の磁気光学カー回 転スペクトルのFe層厚依存性



After Y. Suzuki (AIST)

## Magneto-optical ellipticity at 4eV vs. thickness of Fe layer 4eVにおける1層あたりのカー楕円率のFe層厚依存性



#### Au-thickness dependence of Co/Au/Co Co/Au/Coの磁気光学効果のAu層厚依存性



After Y.Suzuki

### Artificial ordered alloy of Fe/Au Fe/Au人工規則合金

- [Fe(1ML)/Au(1ML)]<sub>N</sub> is a L1<sub>0</sub> type ordered alloy that does not exist in nature (Peritechtic system) [Fe(1ML)/Au(1ML)]<sub>N</sub>は天然には存在しないL1<sub>0</sub>型の規則合金である。
- At interfaces in Fe/Au, L1<sub>0</sub> type Fe(1ML)/Au(1ML) exists
   [Fe(xML)/Au(xML)]<sub>N</sub>においても、Fe層とAu層の界面にはL1<sub>0</sub>型Fe(1ML)/Au(1ML)が存在
- New band structure appears due to hybridization FeとAuの間には電子の混成が生じ、新しいバンド構 造が出現している。

# Atomic arrangement in a unit cell of Fe(1ML)/Au(1ML) with a L1<sub>0</sub> structure Fe(1ML)/Au(1ML)人工規則合金の結晶構造



整数・非整数層厚をもつFe/Au人工格子 Superlattices : [Fe(xML)/Au(xML)]<sub>N</sub> with integer and non-integer layer thickness x=1, 1.25, 1.5, 1.75,1, 2.25, 2.5, 2.75, 3.25, 3.5, 3.75, 4, 6, 8, 10, 15



#### Retardation modulation technique 光学遅延変調法(円偏光変調法)



## Magneto-optical spectrometer system 磁気光学スペクトル測定系



#### Magneto-optical Kerr spectra in Fe/Au superlattices Fe(xML)/Au(xML)人工格子における 磁気光学カー回転角のスペクトル



 $x=1\sim 5$ 

 $x = 6 \sim 15$ 



Calculated Kerr spectra in Fe/Au superlattices using abinitio band calculation 第1原理バンド計算によるFe(xML)/ Au(xML)人工格子の磁気光学スペクトル (山口による)

The structure around 4eV can be assigned to Au(5d  $\downarrow$ ) to Fe(3d  $\downarrow$ ) transition 4eV付近に見られる構造は、Auの 5d  $\downarrow$  バンドからAuの5f  $\downarrow$  バンドへ の遷移である。Auの5f  $\downarrow$  バンドは Feの3d  $\downarrow$  バンドと強く混成してお り、実質的にはAu(5d  $\downarrow$ )→Fe(3d  $\downarrow$ ) 遷移と見なせる。

#### Peak position of Kerr rotation vs. modulation period Fe/Au人工格子の磁気光学スペクトルのピーク位置の変調周 期に対するプロット。



Dotted curve denotes the peak position in magneto-optical spectra of Au/Fe/Au ultrathin trilayer 点線は、超薄膜にお ける量子閉じ込め ピークの変調周期依 存性。

# MSHG study of magnetism for surfaces and interfaces



Magnetic second harmonic generation (MSHG) has been applied to study of magnetic thin film and multilayer

#### Scheme

Fe/Au superlattices with a modulation of mono-atomic layers



MSHG technique was applied to Fe/Au superlattices



• For weak incident laser field  $E(\omega)$ : linear

$$P_i^{(1)} = \chi_{ij}^{(1)} \varepsilon_0 E_j \qquad \text{response}$$

- For strong incident laser field  $\mathsf{E}(\omega)$  :

$$P_{i} = \varepsilon_{0} (\chi_{ij}^{(1)} E_{j} + \chi_{ijk}^{(2)} E_{j} E_{k} + \chi_{ijkl}^{(3)} E_{j} E_{k} E_{l} + \cdots)$$

 Third rank tensor is not allowed in centrosymmetric materials.

Vonlinear

response

• Nonlinear polarization  $P^{(2)}$  for incident field of  $E = E_0 \sin \omega t$ 

$$P^{(2)} = \varepsilon_0 \chi^{(2)} \frac{E_0}{2} + \varepsilon_0 \chi^{(1)} E_0 \sin \omega t - \varepsilon_0 \chi^{(2)} \frac{E_0^2}{2} \cos 2\omega t + \cdots$$

Second harmonic generation (SHG)

### Nonlinear magneto-optical effect measurement system







[Fe(3.5ML)/Au(3.5ML)] superlattice (Sin)

inear Kerr rotation & elliptic  $\theta_{\rm K}^{(2)}$ = 17.2 °  $\eta_{\rm K}^{(2)}$ =3°

### Result

#### Nonlinear Kerr rotation : $\Delta \phi$



Analyzer angle dependence

## Azimuthal angle-dependence of MSHG



Azimthal angle-dependence of MSHG intensity for [Fe(3.75ML)/Au(3.75ML)] superlattice.  $(P_{in} P_{out})$ 





Surface non-magnetic term

•SHG response causes an isotropic contribution only.

Bulk non-magnetic term

$$P_{i}(2\omega) = \chi_{ijk}^{(D)} E_{j}(\omega) E_{k}(\omega) + \chi_{ijkl}^{(Q)} E_{j}(\omega) \nabla_{l} E_{k}(\omega)$$

• For crystallographic contribution the electric quadrupole should be introduced to get four rank tensor.

 $\implies$  SHG response causes an anisotropic contribution (parameter B).

#### Surface magnetization induced term

$$\chi_{ijk}^{s}(M) = \chi_{ijk}^{s}(0) + X_{ijkL}^{s}M_{L}$$

• The surface magnetic response comes from the electric dipole term expanded by magnetization and contributes to the parameter C.

## Calculated azimuthal angle dependence of SHG and MSHG signals

input-output polarization	surface non-magnetic	bulk non-mangetic	surface magnetization-induced	sum
S <sub>in</sub> -S <sub>out</sub>	0	$ B\sin 4\phi ^2$	$ \pm A_{ss}\pm C\cos 4\phi ^2$	$ \pm A_{\rm ss}+B\sin4\phi\pm C\sin4\phi ^2$
S <sub>in</sub> -P <sub>out</sub>	$ \mathbf{A'_{sp}} ^2$	$ A_{sp} - B\cos 4\phi ^2$	$ \pm C\sin 4\phi ^2$	$ A_{\rm sp}-B\cos 4\phi\pm C\sin 4\phi ^2$
P <sub>in</sub> -S <sub>out</sub>	0	$ -B\sin 4\phi ^2$	$ \pm A_{ps}\mp C\cos 4\phi ^2$	$ \pm A_{\rm ps} - B\sin 4\phi + C\cos 4\phi ^2$
P <sub>in</sub> -P <sub>out</sub>	$ A'_{pp} ^2$	$ A_{pp}+B\cos 4\phi ^2$	$ \mp C\sin 4\phi ^2$	$ A_{\rm pp}+B\cos 4\phi \mp C\sin 4\phi ^2$

Kerr rotation calculated from parameters Axx,B,C  $\Theta_{\rm K}^{(2)} = (\psi_+ - \psi_-)/2$ 

$$S_{in} \quad \tan 2\psi_{\pm} = \frac{2(A_{SP} - B\cos 4\phi \pm C\sin 4\phi)(\pm A_{SS} + B\sin 4\phi \pm C\cos 4\phi)}{(A_{SP} - B\cos 4\phi \pm C\sin 4\phi)^2 - (\pm A_{SS} + B\sin 4\phi \pm C\cos 4\phi)^2}$$

$$\mathbf{P}_{in} \quad \tan 2\psi_{\pm} = \frac{2(A_{PP} + B\cos 4\phi \mp C\sin 4\phi)(\pm A_{PS} - B\sin 4\phi \pm C\cos 4\phi)}{(A_{PP} + B\cos 4\phi \mp C\sin 4\phi)^2 - (\pm A_{PS} - B\sin 4\phi \pm C\cos 4\phi)^2}$$



#### Azimuthal angle-dependence of MSHG for a [Fe(3.5ML)/Au(3.5ML)] superlattice (Sin-Pout, Sin-Sout configuration)



The equation of the azimuthal angledependence by theoretical analysis

Sin-Pout

$$I^{SP} = \left| A^{SP} - \underline{B} \cos 4\varphi \pm \underline{C} \sin 4\varphi \right|^2$$

Sin-Sout

$$A^{SS} = \left| \pm A^{SS} \pm \underline{C} \cos 4\varphi + \underline{B} \sin 4\varphi \right|^2$$

A <sup>SP</sup> (surface nonmagnetic term)	= 460
A <sup>ss</sup> (surface nonmagnetic term)	= 100
B(bulk nonmagnetic term)	= 26
C(surface magnetic term)	= -88

#### Calculated and experimental patterns :x=3.5





#### Dots:exp. Solid curve:calc.

A<sup>SS</sup>=100, B=26, C=-88

## The fitting parameter of the azimuthal pattern (Sin-Pout)

$$I^{SP} = \left| A^{SP} - B \cos 4\varphi \pm C \sin 4\varphi \right|^2$$



Modulated rate x (ML) Fig. The fitting parameter of azimuthal angledependence for [Fe(xML)/Au(xML)] ( $1.25 \le x \le 3.75$ ) superlattices.

Contribution of ASP term

- Surface nonmagnetic term
- · Dependence on focused beam power

#### Contribution of **B** term

- · Bulk nonmagnetic term
- The parameter B is constant for the modulation x.

#### Contribution of C term

- Surface magnetic term
- Decrease of the parameter C for the azimuthal patern rotation



## Azimuthal angle dependence of the MSHG intensity for the anlyzer angle

#### Fe(1.25ML)/Au(1.25ML)

Analyzer angle =  $30^{\circ}$ 



Analyzer angle = 60° Analyz (Pin-Pout)



#### Experimental azimuthal angle-dependence of nonlinear Kerr rotation and ellipcity for a Fe(3.75ML)/Au(3.75ML) superlattice.(Sin)



Fig. Calculated azimuthal angle-dependence of nonlinear Kerr rotation  $\theta^{(2)}_{K}$  and ellipcity  $\eta^{(2)}_{K}$  for a Fe(3.75ML)/Au(3.75ML) superlattice.

(a) Nonlinear Kerr rotation

• Azimuthal angle-dependence of nonlinear Kerr ellipticity is found to be sinusoidal.

(b) Nonlinear Kerr ellipticity

$$\eta_{K}^{(2)} = \frac{1}{2} \left[ \tan^{-1} \left( \frac{I_{MAX}(+)}{I_{MIN}(+)} \right) - \tan^{-1} \left( \frac{I_{MAX}(-)}{I_{MIN}(-)} \right) \right]$$

- I: Analyzer angle dependence of the MSHG intensity
- Azimuthal angle-dependence of nonlinear Kerr ellipticity showed 45°-shift compared to Kerr rotation.
- Ellipticity  $\eta^{(2)}_{\kappa}$  was about zero for the maximum  $\theta^{(2)}_{\kappa}$  and the minimum  $\theta^{(2)}_{\kappa}$ .

#### Calculated azimuthal angle-dependence of nonlinear Kerr rotation and ellipcity for a Fe(3.75ML)/Au(3.75ML) superlattice.(Sin)



Azimuthal angle (deg.)

Fig. Experimental azimuthal angle-dependence of nonlinear Kerr rotation  $\theta^{(2)}_{\kappa}$  and ellipcity  $\eta^{(2)}_{\kappa}$  for a Fe(3.75ML)/Au(3.75ML) superlattice.

(a) Nonlinear Kerr rotation  $\theta^{2}_{\kappa}$ 

$$\tan \psi_{\pm} = \frac{2\left(A^{SP} - B\cos 4\varphi \pm C\sin 4\varphi\right)\left(\pm A^{SS} + B\sin 4\varphi \pm C\cos 4\varphi\right)}{\left(A^{SP} - B\cos 4\varphi \pm C\sin 4\varphi\right)^2 - \left(\pm A^{SS} + B\sin 4\varphi \pm C\cos 4\varphi\right)^2}$$
$$\bigoplus \quad \theta_K^{(2)} = \frac{\psi_+ - \psi_-}{2}$$

(b) Nonlinear Kerr ellipticity  $\eta^{\scriptscriptstyle (2)}{}_{\!\scriptscriptstyle \rm K}$ 

 $I^{Sin}(\theta) = |P^{SP}\cos \theta + P^{SS}\sin \theta|^2$ 

I: Analyzer angle-dependence of MSHG intensity for Pin configulation.

$$\eta_{K}^{(2)} = \frac{1}{2} \left[ \tan^{-1} \left( \frac{I_{MAX}(+)}{I_{MIN}(+)} \right) - \tan^{-1} \left( \frac{I_{MAX}(-)}{I_{MIN}(-)} \right) \right]$$



(b) Calculated pattern (Sin)

nonlinear Kerr rotation angle and ellipticity in [Fe(3.75ML)Au(3.75ML)]

#### Experimental and calculated patterns of Kerr rotation angle



Sin configuration: (a) Experimental data,

(b) Calculated using *parameters determined* 

by fitting to the azimuth patterns

## Nonlinear Kerr rotation angle of [Fe(xML)/Au(xML)] $(1.25 \le x \le 3.75)$ superlattices (Sin)



Modulated rate x (ML)

Fig. Nonlinear Kerr rotation angle of [Fe(xML)/Au(xML)] (1.25≤x≤3.75) superlattices [(a)Calculation, (b)Experiment]

#### Calculation and experimental result

Calculated nonlinear Kerr rotation angle  $\theta_{K}^{(2)}$  using the fitting parameter A<sup>SP</sup>, A<sup>SS</sup>, B, C of the azimuthal pattern (The maximum  $\theta_{K}^{(2)}$  was selected for azimuth angle)

- The experimental maximum  $\theta_{K}^{(2)}$  for x=1.75 superlattice was 31.1°.
- The calculated  $\theta_{K}^{(2)}$  reproduced the muximum  $\theta_{K}^{(2)}$  for x=1.75 superlattice.



The nonlinear Kerr rotation was explained by theoretical analysis.

Linear magneto-optical spectra in Co/Ru superlattice Co(5ML)/Ru(5ML)の線形磁 気光学スペクトルの実験値と バンド計算による理論値



Fig. 2: The Kerr (a) and ellipticity (b) spectra of Co(5ML)/Ru(5ML) superlattice. For comparison, those of HCP Co are also shown. Experimental data of HCP Co are in Ref.[14]

### NOMOKE in Co(5ML)/Ru(5ML)



# Azimuthal angle dependence of MSHG in Co(5ML)/Ru(5ML)



## Conclusion

 Magneto-optical spectra in Fe/Cu and Fe/Au system depends strongly on the thickness of the layers in comparison with characteristic length of the material: wavelength of light, de Broglie wavelength of electrons and atomic size. The four-fold pattern clearly reflects the symmetry of the MgO(100) substrate. This suggests that the Fe/Au superlattice is perfectly epitactic to the substrate.

 The azimuthal angle dependence was analyzed in terms of nonlinear electrical susceptibility tensor taking into account the magnetic symmetry of the superlattice.

> •The azimuthal pattern was explained by symmetry analysis, taking into account the surface nonmagnetic A, bulk non-magnetic B and surface magnetic C contributions.

• MSHG was shown to lead to a nonlinear Kerr rotation  $\theta^{(2)}_{K}$ that can be orders of magnitude larger than its linear equivalent (0.2°), e.g.,  $\theta^{(2)}_{K}$  for x=1.75 was 31.1°

 We observed azimuthal angle-dependence of the nonlinear Kerr rotation for the first time.

 The azimuthal angle-dependence of the nonlinear Kerr rotation were explained using parameters determined from azimuthal patterns of MSHG response

Modulation period dependence of parameters:
A (Surface nonmagnetic) is large for short period
B (Bulk nonmagnetic) is nearly constant
C (Surface magnetic) becomes larger with modulation Period.

 Magneto-optical spectra of Co/Ru superlattice are much reduced from those of Co

- This can be explained in terms of electronic hybridization of electrons between Co and Ru
- Surface magnetic effect observed by MSHG is also found to be reduced.

Linear and nonlinear magneto-optical effect offer helpful tools to investigate symmetry and electronic structure of magnetic materials

## Other topics

- Near-field Magneto-optics
- XMCD

#### What is Near Field Optics? 近接場とは



たもう1つの微小物体による散乱光

場

## プローブの高さ制御



## 集光モード(a)と照射モード(b)



## SNOMによる磁気光学測定

- 1991 Betzig: 光ファイバーをテーパー状に細めたプローブで光磁気記録・再生に成功
- 1992 Betzig: 超微細加工した金属細線リングの偏光像
- 多くの研究あるが、高解像度のMO-SNOM像 は得られていない
- 偏光をファイバを通して伝えるのが困難

### **SNOM-AFM**

- SNOM-AFMモードを利用
- はじめ: クロスニコル法→コントラスト比とれない
- ・ 解決法: PEMによる偏光変調
- ファイバー特性の測定とよいプローブの選別
- 偏光伝達特性の補償
- 約0.1µmの解像度を達成

## MO- SNOM (polarization modulation technique)











### 0.2µm マークのトポ像と磁気光学像

解像度の定義



トポ像

MO 像

ラインプロファイル

## X線磁気光学効果



## L吸収端の磁気円二色性



## XMCD顕微鏡



## X線顕微鏡によるMO膜観測



mark/space 0.2/0.20.1/0.10.05/0.05 0.1/0.70.05/0.75 0.8/0.80.4/0.40.2/0.2μm

1µm

## X線顕微鏡で観察したGdFeの磁区







### サニャックSNOM



## ポンププローブ磁気光学測定



### まとめ

- 磁気光学効果の基礎を、電磁気学的アプロー チでのべた。この効果が誘電率テンソルの非 対角成分から生じることがわかった。
- 誘電率テンソルの非対角成分は、量子論に基づいて電子エネルギー準位間の光学遷移により説明できることがわかった。
- 磁気光学スペクトルの実例を示し、それらが、 電子構造から予測可能であることを示した。