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CONDENSED MATTER

Role of Interfaces in the Permittivity Tensor of Thin Layers of a Ferromagnetic Metal

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It is known from experimental studies that the components of the permittivity tensor ε depend on layer thicknesses of multilayer thin films, and for nanometer layers, it is necessary to additionally consider the interlayer interfaces. This study provides an answer to the question of what is the reason for the influence of these interfaces on film properties. It is shown that the contribution of interband matrix elements for ferromagnetic films with off-diagonal components of the permittivity tensor determines the ratio between the diagonal and off-diagonal components of the tensor ε at a ferromagnetic layer thickness of about 10 nm.

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1. It is known that interface phenomena largely determine the properties of various multilayer structures [1, 2]. Interfaces affect the performance of devices fabricated on semiconductor heterostructures [3, 4]. In magnetic spintronic heterostructures, the passage of the spin-polarized current and the magnitude of the giant magnetoresistance effect depend on the properties of the interface between the magnetic and nonmagnetic layers [5–7]. In this study, we consider the influence of interfaces between the magnetic and nonmagnetic layers on the magneto-optical properties of multilayer structures, in particular, on the ratio between the diagonal and off-diagonal components of the permittivity tensor ε.

Spectral magneto-optical ellipsometry studies of the frequency dependence of the components of the permittivity tensor ε of thin iron films in the layered structure Fe layer/artificial SiO_2 oxide/Si (100) substrate [8] revealed that the components of the permittivity tensor ε for thin Fe layers with thicknesses of (77.0 ± 0.6), (33.5 ± 0.6), and (11.5 ± 0.6) nm depend not only on the light frequency but also on the layer thickness. At the same time, the values of the components of the tensor ε for a sample with an iron layer thickness of $d(\text{Fe}) = (160.5 \pm 0.8)$ nm allow one to consider it as a bulk sample [9], i.e., as a sample with a thickness much greater than the skin layer thickness. Moreover, data for the thinnest layers ($d(\text{Fe}) = (11.5 \pm 0.6)$ nm) indicate the need to consider the Fe/vacuum (0.58-nm-thick 50% iron layer) and Fe/SiO_2 (0.12-nm-thick 50% iron layer) interfaces, while for the thicker films, the components of the permittivity tensor are insensitive to the interlayer interfaces. In this regard, the question arises: What is the reason for the influence of interfaces on the thin film properties? Possibly, it is associated with the rearrangement of the electronic structure near the surface or with a change in the matrix elements of dipole interband transitions.

In this work, to answer this question, the general expressions for the real and imaginary parts of the components of the permittivity tensor are analyzed. A simplified representation separating the contributions from the interband density of states and dipole interband matrix elements is obtained, indicating that the ratio of the imaginary parts of the diagonal and off-diagonal components of the tensor is independent of the frequency. Experimental data for thin layers ($d(\text{Fe}) = (11.5 \pm 0.6)$ nm) show that this ratio is, in fact, constant in almost the entire measured spectral range, but varies in a narrow frequency range.

2. The permittivity tensor ε of a magnetized ferromagnet describes induced anisotropy and, when the magnetization vector is parallel to the z axis, has the form [10, 11]

$$[\varepsilon] = \begin{bmatrix} \varepsilon_{xx} & \varepsilon_{xy} & 0 \\ \varepsilon_{yx} & \varepsilon_{yy} & 0 \\ 0 & 0 & \varepsilon_{zz} \end{bmatrix}. \quad (1)$$

When calculating the components of the permittivity tensor $\varepsilon(\mathbf{q}, \omega)$ as functions of the frequency within the linear response theory [12] and the band theory

based on the density functional theory [13], the following expressions are used for the imaginary part of the permittivity tensor at the wave vector $q \to 0$:

$$\mathrm{Im}\varepsilon_{\alpha\beta}(\omega) = \frac{4\pi^2 e^2}{\Omega}\lim_{q\to 0}\frac{1}{q^2}\sum_{c,v,k}2\delta(E_{ck} - E_{vk} - \omega) \times \langle u_{c,k+e_\alpha q}|u_{v,k}\rangle\langle u_{c,k+e_\beta q}|u_{v,k}\rangle^*. \quad (2)$$

Here, the indices c and v denote unfilled and filled bands (for semiconductors, conduction and valence bands, respectively), $u_{\lambda k}$ is the wavefunction of the orbital $\lambda = c, v$ with the wave vector k, Ω is the unit cell volume, e_α are the components of the unit vector, and multiplier 2 appears owing to the summation over the electron spin.

The real part of the permittivity tensor is usually specified by the Kramers–Kronig formula [13]

$$\mathrm{Re}\varepsilon_{\alpha\beta}(\omega) = 1 + \frac{2}{\pi}P\int_0^\infty \frac{\mathrm{Im}\varepsilon_{\alpha\beta}(\omega')\omega'}{\omega'^2 - \omega^2 + i\eta}d\omega'. \quad (3)$$

When calculating the permittivity of a metal, there are bands *d* and *sp* instead of the valence and conduction bands, respectively. Accordingly, the expression for the imaginary part of the permittivity can be rewritten in the long-wavelength limit $q \to 0$ using matrix elements of interband transitions $\langle u_{c,k+e_\alpha q)}|u_{v,k}\rangle = d_{cv}^\alpha(k)$:

$$\mathrm{Im}\varepsilon_{\alpha\beta}(\omega) = \frac{4\pi^2 e^2}{\Omega}\lim_{q\to 0}\frac{1}{q^2}\sum_{c,v,k}2\delta(E_{ck} - E_{vk} - \omega) \times d_{cv}^\alpha(k)d_{cv}^\beta(k)^*. \quad (4)$$

Thus, the expression for the imaginary part of the diagonal component of the tensor $\mathrm{Im}\varepsilon_{xx}$ includes the square of the *x* component of the interband matrix element $|d_{cv}^x|^2$, depending on the wave vector k, and the expression for the imaginary part of the off-diagonal component $\mathrm{Im}\varepsilon_{xy}$ includes the product of the components $d_{cv}^x(d_{cv}^y)^*$.

If the matrix elements weakly depend on the wave vector, then the matrix elements $\langle d_{cv}^x\rangle$ and $\langle d_{cv}^y\rangle$ averaged over the Brillouin zone can be removed from the sum over k in Eq. (4). The interband density of states $N_{cv}(\omega)$ is represented in the form

$$N_{cv}(\omega) = \frac{1}{\Omega}\sum_k 2\delta(E_{ck} - E_{vk} - \omega). \quad (5)$$

Then, the imaginary parts of the tensor components take the form

$$\mathrm{Im}\varepsilon_{xx}(\omega) = 4\pi^2 e^2\sum_{c,v}N_{cv}(\omega)|\langle d_{cv}^x\rangle|^2, \quad (6)$$

$$\mathrm{Im}\varepsilon_{xy}(\omega) = 4\pi^2 e^2\sum_{c,v}N_{cv}(\omega)\langle d_{cv}^x\rangle\langle d_{cv}^y\rangle^*. \quad (7)$$

Such an approximation allows us to separate the contributions from changes in electronic energies caused by interfaces (which are included in the interband density of states), from changes in the matrix elements of interband transitions. We do not know in advance how strongly the matrix elements depend on the band number; in the simplest approximation of two bands ($sp = 1$, $d = 2$), the sum over the band indices is removed, and Eqs. (6) and (7) are represented in the form

$$\mathrm{Im}\varepsilon_{xx}(\omega) = 4\pi^2 e^2 N_{12}(\omega)|\langle d_{12}^x\rangle|^2, \quad (8)$$

$$\mathrm{Im}\varepsilon_{xy}(\omega) = 4\pi^2 e^2 N_{12}(\omega)\langle d_{12}^x\rangle\langle d_{12}^y\rangle^*. \quad (9)$$

It is seen that the main difference between the imaginary parts of the diagonal and off-diagonal components of the tensor ε in Eqs. (8) and (9) is the difference between the matrix elements, while the interband density of states is included in both components equally. Meanwhile, the processing of previous experiments in [8] with and without taking into account the interfaces provides the same diagonal components of the tensor (1) for all thicknesses of iron films. At the same time, the off-diagonal component of the tensor (1) depends on the presence of an interface for the thinnest films. Hence, below we verify the hypothesis that the contribution of the matrix elements in Eqs. (8) and (9) determines the contribution of the interfaces to the off-diagonal components of the permittivity tensor.

According to Eqs. (8) and (9), the ratio $\mathrm{Im}\varepsilon_{xx}(\omega)/\mathrm{Im}\varepsilon_{xy}(\omega)$ is the ratio of the matrix elements $\langle d_{12}^x\rangle/\langle d_{12}^y\rangle$, and it is expected that it does not depend on the energy (and on the possible interface-induced energy changes), which can be verified experimentally.

3. Using the components of the permittivity tensor of the Fe layer obtained in the spectral magneto-optical ellipsometry study of $Fe/SiO_2/Si$ samples prepared by thermal evaporation in vacuum [8, 9], we calculated the dependence of the ratio $\mathrm{Im}\varepsilon_{xx}(\omega)/\mathrm{Im}\varepsilon_{xy}(\omega)$ on the light frequency in the visible spectral range for the thinnest sample with an iron layer thickness of (11.5 ± 0.6) nm (Fig. 1). The components of the tensor ε were calculated using a multilayer sample model and taking into account the Fe/vacuum (0.58-nm-thick 50% iron layer) and Fe/SiO_2 (0.12-nm-thick 50% iron layer) interfaces.

It can be seen that the ratio $\mathrm{Im}\varepsilon_{xx}(\omega)/\mathrm{Im}\varepsilon_{xy}(\omega)$ for this sample is independent of the energy of the incident radiation over most of the visible spectrum. The outlier in the region of 3–3.3 eV is due to the zero crossing of the off-diagonal element $\mathrm{Im}\varepsilon_{xy}(\omega)$. The features of the spectrum in this region were discussed in detail in [8], where the spin-polarized density of electronic states was calculated. According to this calculation (Fig. 3 in [8]), the singularity in Fig. 1 in the

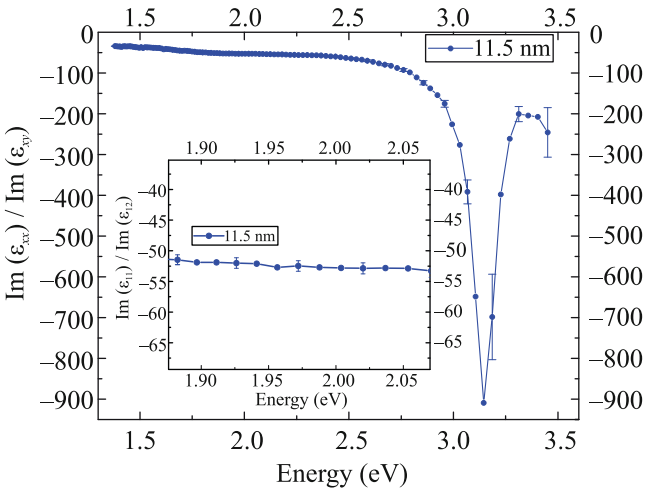

Fig. 1. (Color online) Ratio of the imaginary parts of the diagonal and off-diagonal components of the permittivity tensor of a thin-film $Fe/SiO_2/Si$ sample. The inset shows an enlarged spectral interval of 1.875–2.075 eV.

energy range of 3–3.3 eV is associated with *d–p* interband transitions.

4. As seen in Fig. 1, and especially in the inset, the ratio of matrix elements $\langle d_{12}^x\rangle/\langle d_{12}^y\rangle$, in fact, can be considered constant in almost the entire interval of the measured frequency range. In this interval, the spectral dependence of the tensor components is determined by the interband density of states and it can be assumed that the difference between the diagonal and off-diagonal components is due only to the difference in the matrix elements of interband transitions. At the same time, in the energy range from 3 to 3.3 eV, the ratio of matrix elements sharply increases in absolute value. Comparison in this energy range of the absolute values of the imaginary parts of the diagonal and off-diagonal components of the tensor given in [8] shows that $\mathrm{Im}\varepsilon_{xx}(\omega)$ is small but nonzero, while $\mathrm{Im}\varepsilon_{xy}(\omega)$ is close to zero. This is possible if the matrix element d_{12}^y is close to zero for energies from 3 to 3.3 eV. For thicker samples (with the Fe layer thicker than 33 nm), where the role of the interface is insignificant, $\mathrm{Im}\varepsilon_{xy}(\omega)$ does not vanish and there is no singularity in the ratio $\mathrm{Im}\varepsilon_{xx}(\omega)/\mathrm{Im}\varepsilon_{xy}(\omega)$ in this energy range. Thus, the suppression of the matrix element d_{12}^y for energies from 3 to 3.3 eV for films with a thickness of 11 nm is due to the interface.

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