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To cite this article: E A Vilkov *et al* 2021 *J. Phys.: Conf. Ser.* **2036** 012024

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# THz radiation spectra in magnetic Fe/Mo and Fe/Co<sub>2</sub>FeAl junctions

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**Abstract.** An investigation was made of THz radiation spectra in the Fe/Mo and Fe/Co<sub>2</sub>FeAl magnetic junctions arising from the pumping by a high-density current of the upper spin subband at the Fe/Mo interface. The tensor character of the exchange interaction constant at the Fe/Mo interface, associated with the presence of the DMI-like interaction, promotes radiative relaxation.

## 1. Introduction

At present, the generation of THz radiation in magnetic structures due to spin pumping is of considerable scientific interest [1, 2]. As shown in [3], when a spin-polarized current flows through ferromagnet-ferromagnet contacts, radiative spin-flip transitions occur, and their probability is significantly affected by the anisotropy of the  $sd$  exchange interaction. Shiba [4] was the first to point out the possibility of such anisotropy when considering the Kondo effect in metals with ferromagnetic impurities, presenting the exchange constant in the Heisenberg Hamiltonian as a tensor with nonzero off-diagonal components. In a similar way, one can describe the Dzyaloshinsky-Moriya interaction (DMI) [5, 6], which, in particular, arises at the ferromagnet-heavy metal boundaries [7] and is associated with the manifestation of the effect of spin-orbit interaction. We assume that an anisotropy of the  $sd$  exchange interaction appears in the ferromagnet-heavy metal contact, which leads to the generation of THz radiation when a current is passed through it. In this work, we investigated the THz emission spectra in the Fe/Mo and Fe/Co<sub>2</sub>FeAl magnetic junctions.

## 2. Experiment

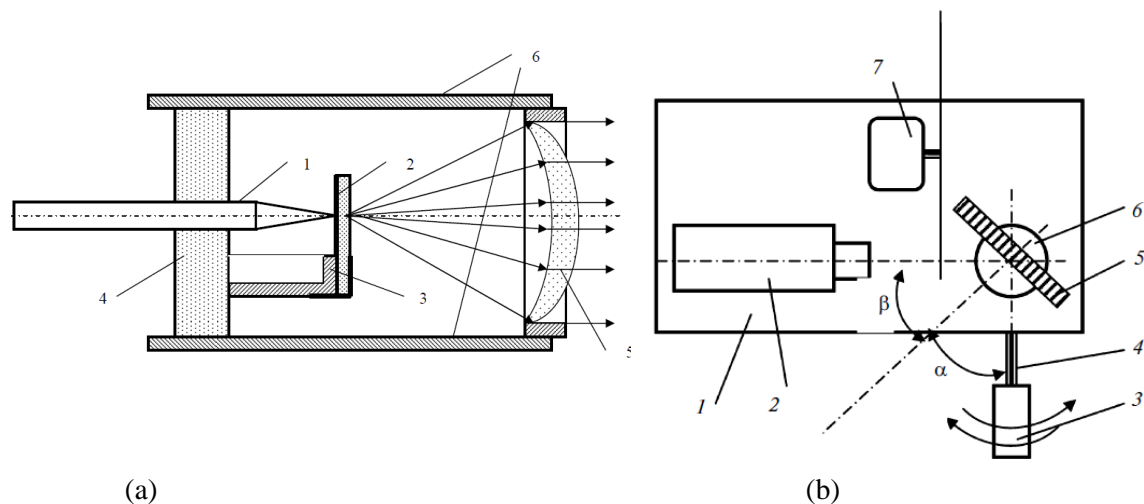
In the experiments, thin films of Mo and Co<sub>2</sub>FeAl Heusler alloy 20 nm and 15 nm thick, grown on the R-plane of sapphire by the method of pulsed laser evaporation in an ultrahigh vacuum were used. An Fe rod with a sharp end 20  $\mu\text{m}$  in diameter was brought to the surface of the film until a good electrical contact was formed (Fig. 1a). When a potential difference was applied between the rod (1) and the substrate holder (3) in contact with the film (2), a current flowed through the contact. According to our estimates, the excitation of THz radiation at the rod-film junction requires a current density of at least  $10^6$  A/cm<sup>2</sup>. The highest current density is achieved in the film along the diameter of the rod tip, where the conducting section is  $S = \pi D \Delta$  ( $D$  is the diameter of the rod tip,  $\Delta$  is the film thickness). The main condition that determines the film thickness is its transparency for THz radiation, that is, it should be less than the skin depth thickness, which for THz frequencies is about 30 nm. Based on this, the diameter of the rod tip is  $D \leq 50$   $\mu\text{m}$ . Magnetic domains in a ferromagnetic rod are aligned along its



axis due to the shape anisotropy. As a result, the electrons carrying the current in the rod are spin polarized along this direction.

The radiation arising in the region of the rod-film contact was focused by a meniscus lens (5) for THz radiation made from a high resistive Si and was directed to a recording detector, an optoacoustic receiver TYDEX OAP-GC1R (Golay cell). The radiation recorded by the detector lies in a wide range of wavelengths, from 10  $\mu\text{m}$  to 8 mm. To cut off the long-wavelength signal, a low-frequency filter in the form of a metal grid with cells of  $125 \times 125 \mu\text{m}^2$  was used. A high-frequency polymer filter TYDEX, which cut off frequencies above 10 THz was also used. Then the signal from the detector was amplified, digitized on an ADC AKTAKOM and fed to a PC. The parameters of the supply voltage and current were also supplied there. Thus, by changing the current, it was possible to see the radiation response in real time.

To study the spectral characteristics of THz radiation, we used a diffraction grating with a step of 20  $\mu\text{m}$ , rotated with respect to the receiver by an angle  $\beta = 30^\circ$ . In this case, the source could rotate with respect to the grating at angles lying in the range  $-90^\circ < \alpha < 40^\circ$ , as shown in Fig. 1b.



**Figure 1.** a) THz emitter with a "rod-film" structure. 1 – ferromagnetic rod, 2 – film on a dielectric substrate, 3 – substrate holder, 4 – dielectric base platform of the emitter, 5 – meniscus focusing lens, 6 – lens holder. Arrows show the radiation flux.

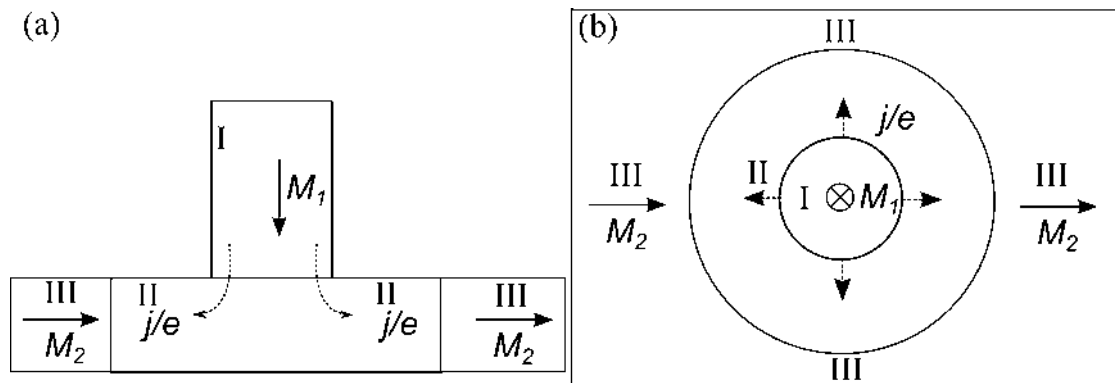
b) The optical system for THz spectral measurements. 1 – optic bench, 2 – THz detector (Golay cell), 3 – THz emitter, 4 – rotary device, 5 – diffraction grating, 6 – diffraction grating holder.

### 3. Theoretical model

Let us consider the model of the "rod-film" magnetic junction (Fig. 2). The Hamiltonian of free electrons in magnetic conductors can be written in the form [2]

$$\hat{H}(\hat{\mathbf{p}}) = \hat{\sigma}_0 \frac{\hat{\mathbf{p}}^2}{2m^*} - \hat{\boldsymbol{\sigma}} \hat{\mathbf{I}}(\hat{\mathbf{p}}) \quad (1),$$

where is  $m^*$  the effective mass of an electron,  $\mathbf{p}$  is the operator of the generalized canonical momentum,  $\boldsymbol{\sigma}$  is the vector of Pauli matrices, and  $\sigma_0$  is the  $2 \times 2$  unit matrix. The electric current in the structure is directed from region I to active region II. The electromagnetic field can be taken into account in the  $s$ - $d$  exchange interaction due to the dependence of the exchange energy on the electron momentum [8]. In the presence of an electromagnetic field, the momentum operator in the formula (1) must be replaced by  $\hat{\mathbf{p}} - (e/c)\vec{A}$ , where  $\vec{A}$  is the vector potential of the external electromagnetic field,  $e$  is the electron charge, and  $c$  is the speed of light in vacuum.



**Figure 2.** Scheme of the magnetic junction: a – side view; b – top view; I – rod; II – injection region of the film (with nonequilibrium spin polarization); III – areas of the film outside the injection

After substitution in the Hamiltonian and linearization with respect to the small parameter  $\bar{A}$ , we obtain

$$\hat{H}(\hat{p}) \approx \hat{\sigma}_0 \frac{\hat{p}^2}{2m^*} - \hat{\sigma}_0 \frac{e}{m^* c} \hat{p} \bar{A} - \hat{\sigma} \bar{I}(\vec{p}) \Big|_{\vec{p}=\hat{p}} - \hat{\sigma} \frac{e}{c} \frac{\partial \bar{I}}{\partial \vec{p}} \bar{A} \Big|_{\vec{p}=\hat{p}} \quad (2).$$

The last term in (2), which has off-diagonal elements, is responsible for the electron spin flip mechanism in interband transitions. The exchange interaction between the conduction electron and the localized electron of the lattice can be represented in the Heisenberg form [9].

$$H_{ex} = -J \mathbf{S}_1 \cdot \mathbf{S}_2 \quad (3).$$

In this case, the anisotropic addition arising from the spin-orbit interaction, in the first approximation, can be represented in a form similar to that of the DMI [7]

$$H_{DM} = D_{12} (\mathbf{S}_1 \times \mathbf{S}_2) \quad (4).$$

Adding the DMI-like term to the Heisenberg Hamiltonian allows us to represent the effective exchange interaction in tensor form

$$\vec{J} = \begin{pmatrix} J & D_3 & -D_2 \\ -D_3 & J & D_1 \\ D_2 & -D_1 & J \end{pmatrix} \quad (5).$$

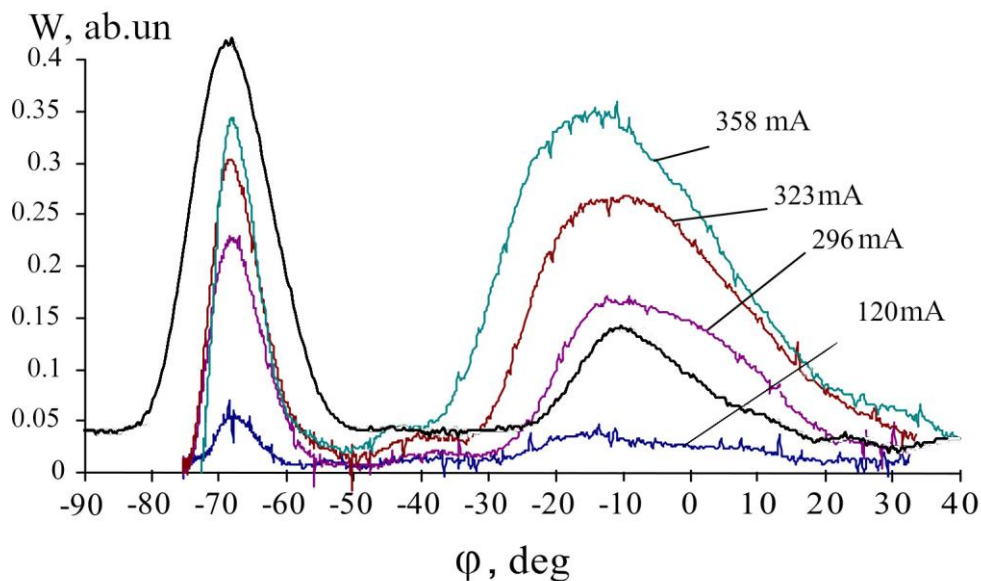
Here  $J$  is the Heisenberg exchange constant,  $D_1$ ,  $D_2$  and  $D_3$  are the components of the Dzyaloshinsky vector. The exchange interaction per unit volume with allowance for anisotropy can be written [3] in the following form

$$\mathbf{I} = \hbar \gamma \frac{\mathbf{H}_{sd}}{2} = \mu_B G(\mathbf{p}) \mathbf{M}_2(\mathbf{r}) \quad (6),$$

where  $G(\mathbf{p})$  is the  $sd$ - exchange tensor,  $\mathbf{M}_2$  is the magnetization in the boundary layer, and  $\mathbf{H}_{sd}$  is the exchange field. According to eqs. (5), (6), the exchange field will have components parallel and perpendicular to the quantization axis specified by the direction of magnetization. The exchange field  $\mathbf{H}_{sd}$  can be represented in the form of two components: longitudinal (along the quantization axis), and transverse. As shown in [3], the presence of the transverse component of the exchange field significantly affects the number of quantum transitions of conduction electrons with spin flip.

### Experimental results

The frequency response of the emitter, measured with a diffraction grating, is shown in Fig. 3. As seen in the figure, the radiation spectrum lies in the frequency range 5 - 12 THz. The maximum signal frequency is  $f = 7.0$  THz. The full width at half maximum of the spectral curve was 2.28 THz. The presented curve is not described by the Planck formula, namely, according to the Planck formula, the observed maximum radiation power should correspond to the heating temperature of the emitter 180 K. In addition, the Planck curve for this temperature is more than an order of magnitude wider. The maximum in the Planck distribution for higher temperatures goes to the region of lower frequencies. All this points to the non-thermal nature of the recorded radiation.



**Figure 3.** Frequency characteristics of a spin-injection emitter for different currents, measured using a diffraction grating. Black curve is for Mo film and colored curves are for  $\text{Co}_2\text{FeAl}$  Heusler alloy film.

### 4. Discussion

To explain the appearance of spin-injection radiation, we assume that induced spin polarization of electrons occurs [9] in Mo near contact with Fe, indicating the ferromagnetic ordering of the spins of Mo atoms propagating to the depth of several atomic layers. However, this distance is sufficient for the subbands with spin down and up to move apart by several tens of meV near the iron boundary, which corresponds to several THz. Because of a strong spin – orbit interaction in molybdenum, the DMI - like  $sd$ - exchange anisotropy arises at the interface with iron [7], and a noncollinear magnetization distribution can be realized. In this case, a high-density spin-polarized current flowing from Fe to Mo will pump electrons into the upper spin subband in the induced ferromagnetism layer. The electrons should relax, and the tensor nature of the exchange interaction constant promotes radiative relaxation. In the structure with the  $\text{Co}_2\text{FeAl}$  Heusler alloy the mechanism of formation of THz radiation is mainly determined by the uniform exchange interaction, but because of the half-metallic band structure of the Heusler alloy the spin polarization is close to unity and so the efficiency of dynamic spin-injection radiation is also expected to be high. The results obtained indicate the spin-injection mechanism of the observed THz radiation in the Fe/Mo magnetic junction. It arises due to the pumping by a high-density current of the upper spin subband in the induced ferromagnetism layer in Mo. In this case, the tensor nature of the exchange interaction at the Fe/Mo interface, associated with the presence of DMI like  $sd$ - exchange anisotropy, promotes radiative relaxation of conduction electrons from the upper spin subband of induced ferromagnetism to the lower one.

## 5. Conclusion

The injection of electrons into the boundary region Fe/Mo and Fe/Co<sub>2</sub>FeAl was considered. The two structures differ in the mechanism for the formation of THz radiation. The features of the formation of the radiation mechanism have little effect on the final radiation spectrum. The results obtained can be used to create coherent and non-coherent THz radiation sources. This is especially important, since until now this frequency range (1-30 THz) remains poorly mastered due to the lack of available simple and reliable sources and receivers of THz signals.

## Acknowledgments

Authors wishing to acknowledge financial support from the State Assignment No. 075-00355-21-00.

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