ISSN 1063-7834, Physics of the Solid State, 2021. © Pleiades Publishing, Ltd., 2021.
Russian Text © The Author(s), 2021, published in Fizika Tverdogo Tela, 2021, Vol. 63, No. 9, pp. 1193–1197.

METALS

Terahertz Radiation in the Fe/Mo Magnetic Transition

E. A. Vilkov[a,] *, S. G. Chigarev[a], I. V. Malikov[b], and L. A. Fomin[b]

[a] *Kotelnikov Institute of Radio Engineering and Electronics, Fryazino branch, Russian Academy of Sciences, Fryazino, Moscow oblast, Russia*

[b] *Institute of Microelectronics Technology and High Purity Materials, Russian Academy of Sciences, Chernogolovka, Moscow oblast, Russia*

**e-mail: e-vilkov@yandex.ru*

Received April 9, 2021; revised April 9, 2021; accepted April 19, 2021

Abstract—The terahertz radiation in the Fe/Mo magnetic transition, which occurs when pumping the upper spin subzone in the Fe/Mo interface with a high-density current, has been investigated. The tensor character of the exchange interaction constant on the Fe/Mo interface associated with the presence of the Dzyaloshinskii—Moriya interaction, contributes to radiative relaxation.

Keywords: terahertz radiation, spin-polarized current, anisotropic exchange interaction, Dzyaloshinskii—Moriya interaction

DOI: 10.1134/S1063783421090432

1. INTRODUCTION

Nowadays, the surface magnetic phenomena at the ferromagnetic—heavy metal interfaces [1, 2] associated with the emergence of the Dzyaloshinskii—Moriya (DM) interaction, introduced by Dzyaloshinskii [3] from phenomenological considerations and obtained by Moriya [4] as an effect of spin-orbital interaction, are of significant scientific interest. These phenomena can be interesting from the practical point of view, for example, for generating terahertz radiation due to the spin-flip transitions of conduction electrons between the subzones with opposite spins [5, 6] similar to those occurring in the ferromagnetic-ferromagnetic contacts in the presence of a spin-polarized current passing through them. In this case, as is shown in [7], the probability of radiative transitions is significantly influenced by the anisotropy of the *sd*-exchange interaction of conduction electrons with *d*-electrons of atoms responsible for lattice magnetism. Usually, it is assumed that the exchange interaction is isotropic, however, this is not necessarily the case for the *sd*-exchange interaction. Shiba [8] was the first to point out the possibility of the anisotropy of the *sd*-exchange interaction when considering the Kondo effect in metals with ferromagnetic impurities. He represented the exchange constant in the Heisenberg Hamiltonian as a tensor with nonzero nondiagonal components, which indicates the possibility of spin-polarization of the electron flow and, thereby, the excitation of electromagnetic oscillations due to spin-flip transitions.

In our earlier works, we searched for the materials and structures in which the terahertz radiation due to the spin-injection mechanism could be clearly distinguished against the thermal background. In particular, in addition to the "rod-film" structure, island films of ferromagnetic metal were used, in which the gaps between the islands were filled with an antiferromagnet [9]. Instead of an antiferromagnet, it would also be possible to use a heavy metal in which there is a strong spin-orbital interaction due to which the anisotropy of exchange interaction can be realized at the boundary with iron. It was planned to produce the structures with Fe/Mo/Fe and Fe/W/Fe transitions, where a thin layer of W or Mo would affect Fe, creating an anisotropic exchange interaction. In Mo, the value of the spin-orbital interaction constant is much smaller than that in W. However, this may be an advantage, since the spin relaxation length in Mo is larger, and the electron, having passed through the Mo layer without losing its polarization, will reach another boundary of Fe; at that, the *s*–*d*-exchange interaction at the Fe/Mo interface will be anisotropic.

It was found in [10] that the DMI constant at the Mo/CoFeB interface is $|D| \approx 0.35$ mJ/m^2, which is several times smaller than the known values 0.6–2.9 mJ/m^2 for the Pt/Co interface (see [10] and references there) and 4 mJ/m^2 for Fe/W(110) [11]. To estimate the DMI constant at the Fe/Mo interface, it is possible to assume that $|D| \approx 0.35$ mJ/m^2, which is by one order of magnitude smaller than that for Fe/W.

At first, it was necessary to verify what would happen if electrons were injected directly from the ferromagnetic into the heavy metal. In this work, the authors investigated a spin-injection emitter with a

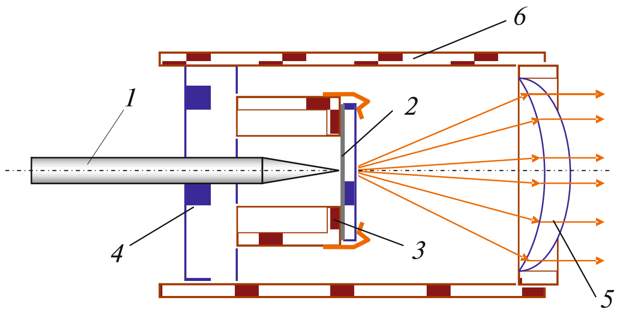

Fig. 1. Spin-injection emitter with a "rod-film" structure. (*1*) the ferromagnetic rod (Fe) with a tip diameter of 10–50 μm; (*2*) the film (Mo) at a dielectric substrate; (*3*) the substrate holder; (*4*) the dielectric base of the emitter; (*5*) the meniscus focusing lens; (*6*) the lens holder. The arrows show the radiation flux.

Fe/Mo magnetic transition. The first results of this investigation are presented and an attempt is made to explain the occurrence of terahertz radiation in such a system.

2. EXPERIMENTAL

In the rod-film radiator [12] (Fig. 1), a rod of Fe with a sharp end with a diameter of 20 μm and films of Mo with a thickness of 20 and 15 nm grown on the R-plane of a sapphire by pulsed laser evaporation in an ultrahigh vacuum were used. The sharp end of the rod was brought to the film surface until getting a good electrical contact between them. When a potential difference was applied between the rod (*1*) and the substrate holder (*3*) contacting with the film (*2*), current was flowing through the contact. According to our estimations, a current density of at least 10^6 A/cm^2 is required to excite terahertz radiation in the Fe/Mo transition. The highest current density is reached in the film over the diameter of the rod tip, where the electrically conducting cross section is $S = \pi D\Delta$ (D is the diameter of the rod tip, Δ is the film thickness). The main condition determining the film thickness is its transparency for terahertz radiation, i.e., it should be smaller than the skin-layer thickness, which is about 30 nm for terahertz frequencies. Based on this, the diameter of the rod tip providing the required current density for excitation of non-thermal radiation, must not exceed $D \leq 50$ μm.

Magnetic domains in the ferromagnetic rod are aligned along its axis due to the anisotropy of its shape. As a result, the electrons that carry the current flowing through the rod are spin-polarized along this direction. The radiation generated in the Fe/Mo contact area was focused by means of a meniscus lens of high-ohmic Si *5* for terahertz radiation and directed to the registering detector—the TYDEX OAP-GC1P (Golay cell) optoacoustic receiver. The used signal receiver is not frequency selective and is capable of detecting radiation in a wide wavelength range from 10 μm to 8 mm. To cut off a long-wave signal, a low-frequency filter in the form of a metal grid with 125 × 125-μm cells was used. The TYDEX high-frequency polymer filter was also used for cutting off the frequencies above 10 THz. Then, the signal from the detector was amplified, digitized at the analog-to-digital converter AKTAKOM and transmitted to a personal computer. The parameters of the supply voltage and current were also recorded there. Thus, by varying the current, it was possible to see in real time the response of the radiation-forming structure to this change in the current. The signal spectrum was investigated by means of a 40-μm diffraction grating. The spectral measurement technique is described more in detail in [13].

3. RESULTS

Figure 2 shows the time dependencies of the jump-wise change in the current flowing through the specimen and the radiation power recalculated to microwatts recorded by the Golay cell for the analyzed terahertz emitter. Analyzing the character of the change in the radiation power due to the jump-wise change in the current shown in Fig. 2a, it is possible to distinguish two following regions: a region with a smooth increase in the power and a region with a sharp change in the power.

It should be noted that the jump-wise character becomes increasingly pronounced with growing current. This gives grounds to our assumption that the steepness of the increase in the power under study is related to the current density. At low currents of up to 100 mA (which corresponds to a current density j lower than 10^7 A/cm^2), the jumps are practically absent. If this value is exceeded, they appear and prevail at the values of current above 200 mA ($j = 2.5 \times 10^7$ A/cm^2). Based on the results previously obtained, we can assume that nonthermal radiation providing a power jump occurs at a current density of higher than 10^7 A/cm^2. An alternative to nonthermal radiation in the terahertz range is thermal radiation whose power increases relatively slowly as the emitter gets heated (the region of smooth power increase in Fig. 2a). Thus, when a current density value of 10^7 A/cm^2 is exceeded, the thermal and dynamic components of the signal can be clearly separated, according to the character of their change in time. To verify this assumption, an experiment was carried out with an emitter providing only thermal radiation. Its tip diameter was 90 μm at which the current density decreased by more than 5 times. At that, even at a current of 200 mA, the current density did not exceed its initial value ~10^7 A/cm^2. To increase radiator heating, the film thickness was reduced down to 15 nm. As a result, we observed only thermal radiation. Its time dependence is shown in Fig. 2b. Based on it, we determined the presence of nonthermal (dynamic) radiation

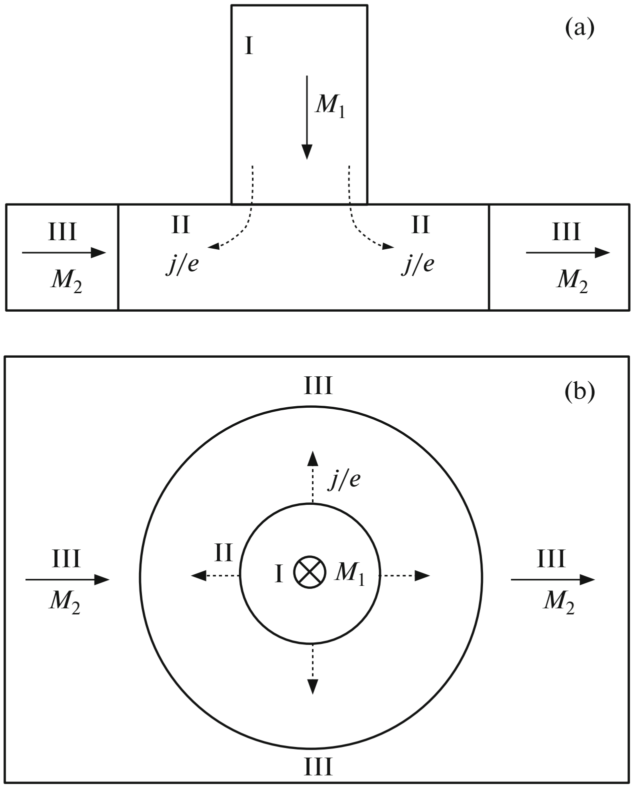

Fig. 2. Time dependence of the radiation power W (curve *1*) in the case of a jump-wise change in the current *I* through the specimen (curve *2*) at a Mo film thickness of 20 nm and a rod tip diameter of 15 μm (a) and at a film thickness of 15 nm and a tip diameter of 90 μm (b).

according to a jump-wise increase in the signal amplitude with a sharp change in the current. So, at a current density higher than 10^7 A/cm^2, it is possible to distinguish two regions characterizing different nature of radiation on curve *1* of Fig. 2: a region of relatively fast growth of the amplitude (dynamic radiation) and a region of its smooth growth (thermal radiation). This allows us to say that in the Fe/Mo transition, nonthermal spin-injection radiation occurs. In the performed experiment, at current of 480 mA, the power $W \approx 60$ μW focused by means of a lens in the paraxial bunch is recorded.

Figure 3 shows the amplitude-frequency characteristic of the radiator measured by means of the diffraction grating. Since when using a diffraction grating, the radiation power is distributed over the diffraction spatial harmonics of the same frequency, the amplitude of the first harmonic shown in Fig. 3, according to which the radiation frequency is estimated, does not correspond to the real radiation power. Therefore, in Fig. 3, the value of radiation power is represented in the units of the signal taken from the GC-1P receiver, i.e., in millivolts. It is seen from Fig. 3 that the radiation spectrum lies within a frequency range from 5 to 12 THz. The maximum signal frequency is f =

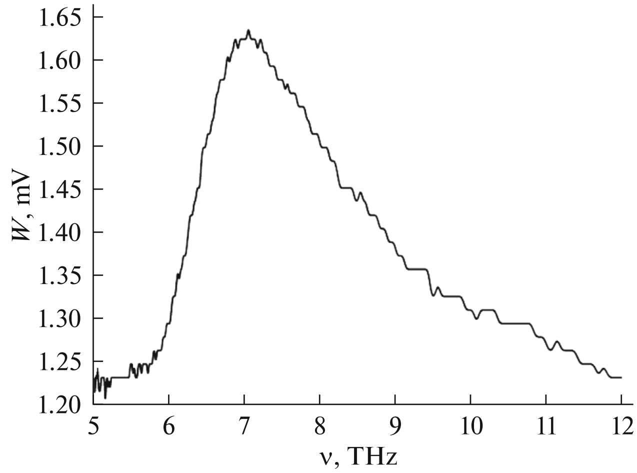

Fig. 3. Amplitude-frequency characteristic of the spin-injection emitter.

7.0 THz. The spectral curve width at a level of 3 dB was 2.28 THz.

Taking into account the diffraction spreading of the peak, this value should be considered as the bounded above assumption. The presented curve is not described by the Planck formula, since, according to the Planck formula, the observed maximum radiation power must correspond to the emitter heating temperature of 300 K. In addition, the Planck curve for this temperature is wider by more than one order of magnitude. The maximum in the Planck distribution for higher temperatures lies in the region of lower frequencies. All this is evidence of a nonthermal character of the detected radiation.

4. DISCUSSION

Due to the presence of the DM interaction at the boundary for the system of bound electrons, the magnetization distribution will be inhomogeneous, which will lead to an anisotropic exchange interaction between the conduction and bound electrons of molybdenum with induced magnetization [7, 14]. The anisotropy of the exchange interaction means that the exchange interaction constant becomes a tensor that may have nondiagonal components. The magnitude of these components will be related to the inhomogeneity of magnetization, namely, to the transverse component of magnetization with respect to the direction of the quantization axis of the system, which, in its turn, is determined by the DM interaction constant. Therefore, to describe the nondiagonal components of the exchange interaction between the conduction and bound electrons, we can, by analogy with the system of bound electrons, write a Hamiltonian in the form of a DM interaction, however, introducing another constant. Let us denote it by D_{sd}. The homogeneous exchange interaction between a conduction electron

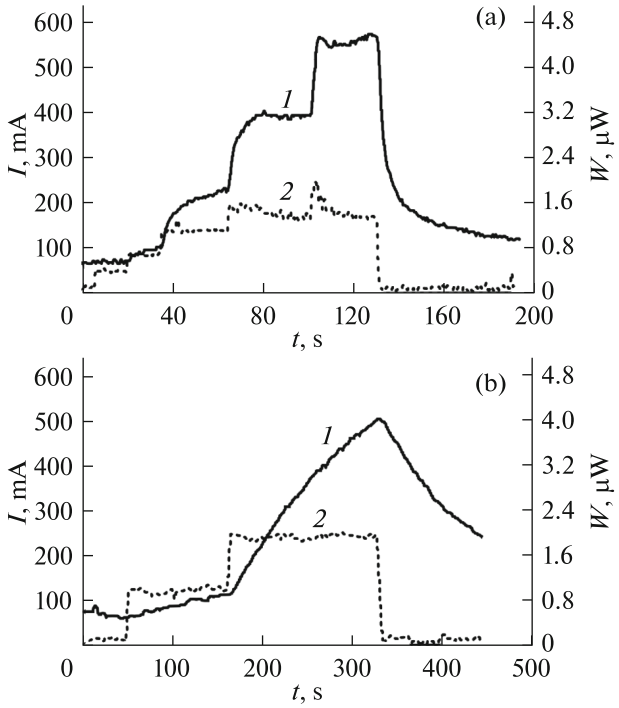

Fig. 4. Time dependence of the radiation power W (curve *1*) in the case of a jump-wise change in the current *I* through the specimen (curve *2*) at a Mo film thickness of 20 nm and a rod tip diameter of 15 μm (a) and at a film thickness of 15 nm and a tip diameter of 90 μm (b).

and a localized electron of the lattice can be represented as follows [15]:

$$H_{\text{ex}} = -J\mathbf{S}_1 \cdot \mathbf{S}_2. \quad (1)$$

As was mentioned above, by analogy with the system of bound electrons, we can represent the anisotropy of the exchange interaction between the conduction and bound electrons in the following form [1]:

$$H_{\text{DM}} = D_{sd}(\mathbf{S}_1 \times \mathbf{S}_2). \quad (2)$$

Adding the DM term to the Heisenberg Hamiltonian allows us to represent the effective exchange interaction in the tensor form as follows:

$$\ddot{J} = \begin{pmatrix} J & D_{sd3} & -D_{sd2} \\ -D_{sd3} & J & D_{sd1} \\ D_{sd2} & -D_{sd1} & J \end{pmatrix}, \quad (3)$$

where J is the Heisenberg exchange interaction constant, D_{sd1}, D_{sd2}, and D_{sd3} are the components of the Dzyaloshinskii vector for the interaction between the conduction and bound electrons. Here, we act by analogy with [3], where it was shown that consideration of the spin-orbital interaction in the first-order approximation gives an addition to the exchange interaction in the form (4); in the higher-order approximations, the exchange interaction constant becomes a tensor of general form. As is shown in [7], the presence of the transverse component of the exchange field causes a significant effect on the number of quantum spin-flip transitions of conduction electrons. If there is a transverse component of the exchange field, the transition probability (the number of transitions per unit time) increases by three orders of magnitude.

To explain the occurrence of spin-injection radiation, we assume that the induced spin polarization of electrons occurs in Mo near the contact with Fe [16]. In [14], generation of terahertz radiation due to the transition of conduction electrons between spin subzones in the noncollinear ferrimagnet with a helicoidal magnetic structure was considered. At that, the electrons with a nonequilibrium spin polarization should be pumped into it from a ferromagnetic injector. The connection of spin and orbital degrees of freedom in the proposed system is attained due to spatially inhomogeneous noncollinear distribution of magnetization. Owing to this, the *sd*-exchange interaction constant will depend on the quasi-momentum of *d*- or *f*-electrons.

We consider electron injection into the Fe/Mo boundary region. In Mo, there is a strong spin-orbital interaction, as a result of which the DM-type interaction of conduction electrons occurs at the Fe/Mo interface [1, 17] and a noncollinear magnetization distribution can also be realized. It is clear, first, that electrons are injected into both spin subzones of the “working” ferromagnet (Mo) even in the case where the magnetizations of both ferromagnets are parallel (the collinearity of the Mo magnetization and the Fe magnetization may be absent because of the presence of DMI; hence, additional investigations are needed). However, in the case of the parallel orientation of magnetization of ferromagnets, a greater amount of electrons “will be injected into the lower spin subzone than into the upper spin subzone”. But even in this case, owing to the spin relaxation to the equilibrium state [18] (the tensor character of the exchange interaction constant contributes to this process), spontaneous photon emission occurs. A thinner film does not exhibit the jump-wise increase in the radiation power not only because of an insufficient current density (as was mentioned above), but also because of the fact that, in our opinion, at small thicknesses, a very sharp increase (by 2–3 times [19]) in the film resistivity occurs even at room temperature. As a result, the film is heated much more intensively and the contribution of spin injection is not noticeable against the background of thermal radiation. To verify this hypothesis, further investigations are needed.

5. CONCLUSIONS

The results obtained point out that the observed terahertz radiation in the Fe/Mo magnetic transition has a nonthermal spin-injection mechanism. It occurs due to the pumping of the upper spin subzone in the boundary Fe/Mo layer with a high-density current. In this case, the tensor character of the exchange interaction at the Fe/Mo interface associated with the presence of the DM interaction contributes to the radiative relaxation of conduction electrons from the upper spin subzone of induced ferromagnetism to the lower spin subzone. The results obtained can be used to create coherent and noncoherent sources of terahertz radiation. This is especially relevant, since until now, this frequency range (1–30 THz) remains rarely used because of the absence of accessible, simple, and reliable sources and receivers of terahertz signals.

FUNDING

The experimental part of this work was partly supported by the Russian Foundation for Basic Research (projects nos. 19-29-03015 mk, 18-29-27020 mk, 20-07-00349A) and also carried out within the framework of the governmental task of IMT RAS no. 075-00355 21-00 and the task of the Kotelnikov Institute of Radio Engineering and Electronics, Fryazino branch, Russian Academy of Sciences. The theoretical part of this work was carried out within the framework of the project of Russian scientific foundation no. 19-19-00607.

CONFLICT OF INTERESTS

The authors declare that they have no conflicts of interest.

REFERENCES

1. A. Fert, Mater. Sci. Forum **59–60**, 439 (1990).
2. X. Z. Yu, Y. Onose, N. Kanazawa, J. H. Park, J. H. Han, Y. Matsui, N. Nagaosa, and Y. Tokura, Nat. Lett. **465**, 901 (2010).
3. I. A. Dzyaloshinsky, J. Phys. Chem. Solids **4**, 241 (1958).
4. T. Moriya, Phys. Rev. **120**, 91 (1960).
5. A. Kadigrobov, Z. Ivanov, T. Claeson, R. I. Shekhter, and M. Jonson, Europhys. Lett. **67**, 948 (2004).
6. Yu. V. Gulyaev, P. E. Zil'berman, A. I. Panas, and E. M. Epshtein, JETP Lett. **86**, 328 (2007).
7. E. A. Vilkov, G. M. Mikhailov, S. A. Nikitov, A. R. Safin, M. V. Logunov, V. N. Korenivskii, S. G. Chigarev, and L. A. Fomin, Phys. Solid State **61**, 941 (2019).
8. H. Shiba, Prog. Theor. Phys. **43**, 601 (1970).
9. E. A. Vilkov, Yu. V. Gulyaev, P. E. Zil'berman, I. V. Malikov, G. M. Mikhailov, A. I. Panas, A. V. Chernykh, and S. G. Chigarev, J. Commun. Technol. Electron. **60**, 1044 (2015).
10. T.-Y. Chen, H.-I Chan, W.-B. Liao, and C.-F. Pai, Phys. Rev. Appl. **10**, 044038 (2018).
11. M. Heide, G. Bihlmayer, and S. Blugel, Phys. Rev. B **78**, 140403(R) (2008).
12. Yu. V. Gulyaev, P. E. Zil'berman, E. M. Epshtein, et al., RF Patent No. 2464683 (2012).
13. S. G. Chigarev, I. V. Malikov, and G. M. Mikhailov, J.Commun. Technol. Electron. **58**, 238 (2013).
14. E. A. Karashtin, JETP Lett. **112**, 122 (2020).
15. S. V. Vonsovskii, *Magnetism* (Wiley, New York, 1971).
16. Y. Murakami, D. Fuse, Y. Sakamoto, Y. Hamada, T. Ono, S. Kitao, M. Seto, M. Tanigaki, T. Saito, and Y. Ohkubo, Hyperfine Interact. **179**, 57 (2007).
17. A. Fert and P. M. Levy, Phys. Rev. Lett. **44**, 1538 (1980).
18. E. A. Vilkov, G. M. Mikhailov, S. G. Chigarev, Yu. V. Gulyaev, V. N. Korenivskii, S. A. Nikitov, and A. N. Slavin, J. Commun. Technol. Electron. **61**, 995 (2016).
19. I. V. Malikov and G. M. Mikhailov, J. Appl. Phys. **82**, 5555 (1997).

Translated by E. Smirnova