

Short Communication

Justification of Visualization Technique of Domain Structure with Raman Scattering

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Qualitative mechanism in line with experimental data on visualization of the domain structure and fine structure of the domain wall in weak ferromagnets has been proposed. The mechanism is based on the phenomenological consideration of Faraday rotation, optical absorption, and atom polarization in response to the radiation exciting Raman scattering. Qualitative agreement of estimates on the scattered radiation intensity in oppositely-magnetized domains with experimental results is good, which made it possible to attack problems of visualization of magnetic entities with nanoscale resolution.

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The intensity of the inelastically scattered light is a function of both polarizability and the number of molecules (elementary cells) within the scattering area. One of the critical drawbacks of Raman scattering is its low effectivity, which amounts to only $IMRS / I_0 \sim 10^{-6}$ of the exciting intensity (I_0) [1], which gives reason to believe the effect of Raman scattering is low. The small probability of Raman scattering per one molecule makes its observation a rather difficult task, which requires advanced hardware. Therefore, the factors, on which the intensity of the Raman signal depends, can be divided into two classes, namely, hardware and natural. The hardware part includes the power and the frequency of the exciting radiation, geometry of the experiment, numerical aperture of the lens, and the detector sensitivity. To the natural factors belong the parameters of the medium studied, namely, polarizability of molecules and atoms ($\vec{P} = \varepsilon \vec{E}$), the absorption coefficient on wavelengths of exciting and scattered radiation, optical activity, and magnitudes of electro- and magneto-optical parameters.

Earlier in the experiment on visualization of the domain structure in a plane sample (100 μm) cut out perpendicular to the optic axis of yttrium orthoferrite (YFeO₃) the difference in the intensity of Raman scattering (34 %) in domains with oppositely oriented magnetic moments was observed (Fig. 1). The present paper proposes qualitative explanation of the observation of domain structure and domain wall with the help of hyperspectral distribution of intensity variations on one line of the Raman spectrum (221 cm^{-1}) proposed in [2].

In analyzing we take into consideration that the exciting (ω_L) and scattered (ω_R) electromagnetic waves are distinct in frequency $\omega_R = \omega_L \pm \omega_k$, where ω_k is the natural frequency of one of possible oscillations in the medium studied. Difference in frequency between exciting and scattered waves allows one to consider them individually. Each follows both absorption laws and Faraday rotation.

Consider the scattered radiation, which was recorded by a spectroscopic camera CCD 1024 \times 256 pixel (pixel size 26 μm).

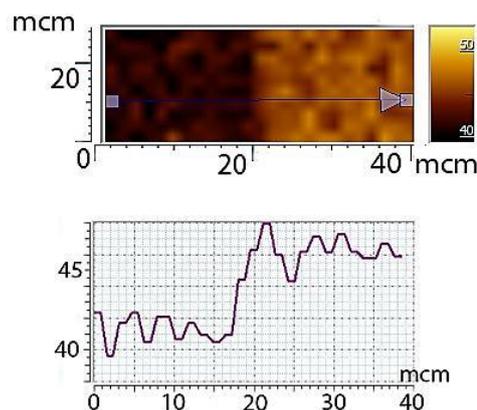


Fig.1 – Hyperspectral distribution of Raman scattering in domains with oppositely-oriented magnetization and within domain wall (top), and its profile (bottom)

Due to Faraday rotation characteristic of YFeO₃ (on $\lambda = 532 \text{ nm}$, $\theta_F = 3000 \text{ }^\circ/\text{cm}$) the electric strength vector of the scattered light rotates through a mirror-symmetric angle in domains with oppositely oriented magnetizations. To the Glan prism, which was an analyzer, light landed from various domains with distinct polarizations. According to Malus's law $I = I_0 \cos^2 \omega$, where I_0 and I are the incoming and outgoing intensities, respectively, the outgoing intensity from variously-magnetized domains is not changed. In other words, the Faraday rotation of scattered light does not contribute to the intensity variations of Raman scattering in oppositely-magnetized domains.

Let us analyze variations in the orientation of the electric strength of the exciting radiation. Geometry of the experiment was not changed for a sample with variously-magnetized domains. The observed varia-

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tions in the Raman intensity in neighboring domains can be described with a ratio W_2/W_1 , where W_2 and W_1 are the Raman intensity in the first and second domains, respectively, which in classical approximation are determined by the magnitudes of projection of the E -vector of exciting radiation onto the polarization vector \vec{P} of atoms. The vector diagram (Fig. 2) illustrates those changes of vectors \vec{E}_0 , \vec{E}_1 and \vec{E}_2 . The rotation of \vec{E}_0 is due to the Faraday rotation

$$\varphi = \theta_F d, \tag{1}$$

where φ is the rotation angle of vector, θ_F is specific Faraday rotation, d is the radiation penetration depth into a sample.

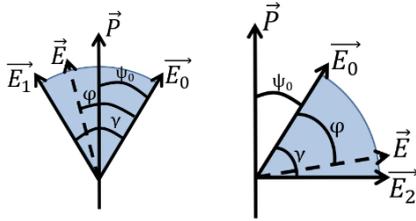


Fig.2 – Vector diagram of mutual orientation of \vec{E} and \vec{P} vectors: left and right for domains with opposite magnetization

To the first approximation anti-Stokes components W_2 and W_1 are determined by expression [3]:

$$W = A(\omega_L - \omega_k)^4 \varepsilon^2 \vec{E}_0^2, \tag{2}$$

where A is a constant; ω_L и ω_k are the frequencies of the exciting radiation and atom natural vibrations, respectively; ε is polarizability; \vec{E}_0 is the electric field vector.

Consider the effect of optical absorption on the exciting intensity. To pinpoint the limiting penetration depth of exciting radiation into a sample, when the Raman signal is still recorded, the following experiment was conducted. Exciting laser (532 nm, 50 mW) radiation was focused on the sample surface. With a neutral slackening filter the laser intensity was decreased with a step of 0.5 dB. The dependence obtained

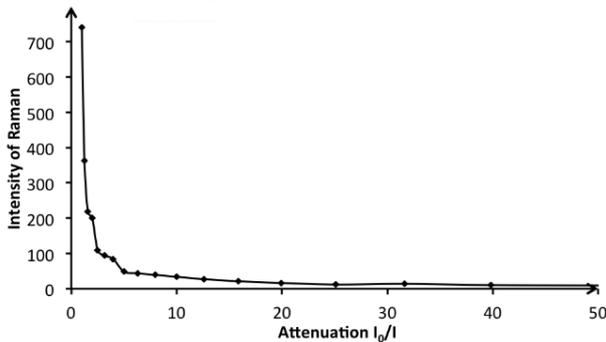


Fig.3 – Effect of optical absorption on the amount of Raman scattering

(Fig. 3) made it possible to establish the fact that the minimum intensity of the exciting radiation I , at which

the Raman recording with the use of the hard-and-software complex was possible, reached $I_0/I = 50$.

Therefore, the penetration depth of exciting radiation can be assessed according to the Lambert-Bouguer law

$$d_0 = -\ln(I/I_0) / \alpha \tag{3}$$

Substitution of the optical absorption coefficient α , whose value for $YFeO_3$ at $\lambda = 532$ nm is 200 cm^{-1} [4], into (3) gives d_0 on the order of $20 \mu\text{m}$. With the use of this d_0 we obtain, from expression (1), the rotation angle γ (Fig. 2) of the vector \vec{E} dictated by the Faraday rotation. In this case, for neighboring domains $\gamma = \pm 60^\circ$.

The angle $\psi_0 = \vec{P} \wedge \vec{E}_0$ (Fig. 2), at which the resulting projections of \vec{E}_1 or \vec{E}_2 onto \vec{P} vary under the influence of Faraday rotation in such a way that when the projection of \vec{E}_1 is maximum, the projection of \vec{E}_2 satisfies the condition $\vec{E}_1 \text{ lim} > \vec{E}_2 \text{ lim}$. To calculate the maximum value of the resulting projection of \vec{E}_1 onto \vec{P} one may use the following equation

$$\frac{d}{d\psi_0} \int_{\psi_0}^{\psi_0 - \gamma} E_0 \cos \psi \cdot d\psi = 0 \tag{4}$$

where τ is the variable angle, considering the Faraday rotation. Extreme of the integration (4) provides $\psi_0 = \gamma / 2 = 30^\circ$.

The exciting radiation power (50 mW) suffices to depolarize atoms. In other words, at the sample surface, when the incident power is maximum, the polarizability vector turns to be parallel to the \vec{E}_0 -vector – $\vec{P} \parallel \vec{E}_0$. As the incident radiation penetrates the matter its power drops and, as a consequence, its influence on the polarizability vector decreases and this vector tends to take its initial position. Under these conditions, in the first domain vectors \vec{P} and \vec{E} rotate with different velocities in one direction, and in the second one they do in mutually opposite directions, which brings about the decrease in Raman intensity.

Expression (3) and dependence in Fig. 3 allow one to evaluate the penetration depth d_1 of the incident radiation (I_0) at which its depolarization effect terminates. In doing so, vector, due to Faraday rotation, rotates through $\varphi_1 = \theta_F d_1$. From Fig. 2 the angle $\psi = (\vec{P} \wedge \vec{E})$ is found in the following way:

$$\psi = \pm \theta_F d + \beta e^{-\alpha d} \tag{5}$$

where β is the angle of polarization variation. Here the first term corresponds to the rotation of the incident strength vector for variously-magnetized domains. The second term is relevant to the rotation of the atom polarizability vector; polarizability decreases with an increase in penetration depth.

To evaluate the observed variations in Raman intensities in neighboring domains characterized by expression W_2/W_1 , we make use of (2), which takes the form for neighboring domains

$$\frac{W_2}{W_1} = \frac{E_0 \left[1 - \int_{\psi_0}^{\psi_0 + \psi_1} (\cos \psi) d\psi + \int_{\psi_0 + \psi_1}^{\psi_0 + \gamma} (\cos \psi) d\psi \right]}{E_0 \left[1 - \int_{\psi_0 + \psi_1}^{\psi_0} (\cos \psi) d\psi + \int_{\psi_0 - \gamma}^{\psi_0 - \psi_1} (\cos \psi) d\psi \right]} \quad (6)$$

Here ψ_1 is determined from (5) after substitution of d_1 . For calculated data, according to (6), we get $W_2 / W_1 = 0.356$, which agrees well with obtained difference in scattered radiation intensities of 0.34 [2]. Some difference can be attributed to the fact that the

real-world crystal structure is not perfect and there is, in this geometry, birefringence, which stems from the error in the sample orientation perpendicular to the optic axis (52° from [001]).

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