New types of surface waves in magnetoelectric antiferromagnets

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The spectrum of the surface polaritons in tetragonal magnetoelectric antiferromagnets is investigated theoretically. It is shown that consideration of the magnetoelectric coupling introduces dispersion of the magnetic permeability in the optical frequency range. This leads to new types of surface modes, i.e., magnetic optical polaritons. The magnetoelectric interaction also results in the appearance of nonreciprocity, i.e., a difference between the rates of propagation of the surface waves along the wave vector and in the opposite direction. The surface modes are always virtual when the wave number is negative. © 1996 American Institute of Physics. [S1063-7761(96)02702-4]

1. INTRODUCTION

Since the discovery of the large value of the magnetoelectric susceptibility in terbium phosphate,¹ there has again been heightened interest in the investigation of various physical properties of magnetoelectric antiferromagnets. In particular, the influence of the magnetoelectric interaction on the spectrum of elementary bulk excitations (magnetoelastic waves) has been the subject of some recent studies.^{2,3}

It would be interesting to investigate the influence of the magnetoelectric effect on the properties of surface waves in antiferromagnets. From the practical standpoint, this interest arises because such modes can be excited by both magnetic and electric fields.

This paper focuses on the spectrum of surface electromagnetic waves (polaritons) in tetragonal magnetoelectric antiferromagnets when a surface wave parallel to the antiferromagnetism vector. It is shown that the magnetoelectric effect results in the appearance of new types of surface polaritons, nonreciprocity, and the conversion of real polaritons into virtual polaritons in the antiferromagnet.

Let us consider a tetragonal antiferromagnet occupying the half-space x>0. We write the energy density of the antiferromagnet in the form^{4,5}

$$F = \frac{1}{2} A \mathbf{L}^{2} + \frac{1}{4} B \mathbf{L}^{4} + \frac{1}{2} a \mathbf{M}^{2} + \frac{1}{2} D (\mathbf{M} \cdot \mathbf{L})^{2}$$
$$+ \frac{1}{2} D' \mathbf{M}^{2} \mathbf{L}^{2} - \mathbf{M} \cdot \mathbf{H} + \frac{1}{2} \beta_{1} (L_{x}^{2} + L_{y}^{2}) + \frac{1}{4} \beta_{2} L_{x}^{2} L_{y}^{2}$$
$$+ \frac{1}{2} \kappa_{\perp}^{-1} (P_{x}^{2} + P_{y}^{2}) + \frac{1}{2} \kappa_{\parallel}^{-1} P_{z}^{2} - \mathbf{P} \cdot \mathbf{E} + F_{ME}, \qquad (1)$$

where A, B, a, D, D', and β_i are parameters of the exchange interaction and the anisotropy, M and L are the ferro- and antiferromagnetism vectors, H and E are the magnetic and electric field strengths, κ_{\perp} and κ_{\parallel} are the dielectric constants, and P is the polarization vector. The term describing the inhomogeneous exchange interaction has been omitted in (1). This enables us to neglect the spatial dispersion of the dynamic magnetic permeability and dielectric constant of the antiferromagnet. The influence of the spatial dispersion of these parameters on the spectrum of the surface modes will be discussed at the end of the paper.

The energy of the magnetoelectric interaction F_{ME} in (1) can be written as⁵

$$F_{ME} = -\gamma_{\alpha\beta\gamma} M_{\alpha} L_{\beta} P_{\gamma}.$$
⁽²⁾

The form of the magnetoelectric coefficient tensor $\hat{\gamma}$ is determined by the specific magnetic symmetry of the crystal. A table of the nonzero components of $\hat{\gamma}$ for crystals of different magnetic symmetries is presented in the appendix.

For tetragonal crystals with a $4z^{\pm} 2x^{-} I^{-}$ structure (Turov's notations^{6,7}) the energy (2) has the form

$$F_{ME} = -\gamma_1 M_z (L_x P_x \pm L_y P_y) - \gamma_2 P_z (M_x L_x \pm M_y L_y)$$
$$-\gamma_3 L_z (M_x P_x \pm M_y P_y) - \gamma_4 M_z P_z L_z, \qquad (3)$$

and for tetragonal crystals with a $4z^{\mp} 2x^{+} I^{-}$ structure this energy is written in the following form

$$F_{ME} = -\gamma_1 M_z (L_x P_y \pm L_y P_x) - \gamma_2 P_z (M_x L_y \pm M_y L_x)$$

$$-\gamma_3 L_z (M_x P_y \pm M_y P_x).$$
(4)

We choose the state with $L_0 || Z$, $M_0 = 0$, and $P_0 = 0$ as the ground state. This state is possible when there are no constant magnetic or electric fields: H_0 , $E_0 = 0$.

To find the spectrum of surface waves we start out from a system of the equations of motion of the magnetization (Landau-Lifshitz) and polarization, as well as Maxwell's equations

$$\dot{\mathbf{M}} = g(\mathbf{M} \times \mathbf{H}_{M} + \mathbf{L} \times \mathbf{H}_{L}) + g\lambda L\mathbf{H}_{M},$$

$$\dot{\mathbf{L}} = g(\mathbf{M} \times \mathbf{H}_{L} + \mathbf{L} \times \mathbf{H}_{M}) + g\lambda L\mathbf{H}_{L},$$

$$\bar{\mathbf{P}} = f\mathbf{H}_{P},$$

$$\operatorname{curl} \mathbf{H} = \frac{1}{c} \frac{\partial \mathbf{D}}{\partial t}, \quad \operatorname{curl} \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t},$$

(5)

div
$$\mathbf{D} = 0$$
, div $\mathbf{B} = 0$

Here g is the gyromagnetic ratio, c is the speed of light in a vacuum, $f \sim z^2/mv_0$, z and m are the charge and reduced mass of a unit cell with a volume v_0 , λ is the damping parameter, which determines the longitudinal relaxation in the magnetic subsystem, and $\mathbf{H}_a = \partial F/\partial a$ ($a = \mathbf{M}$, \mathbf{L} , \mathbf{P}). It was assumed in writing Eqs. (5) that the antiferromagnet is an insulator, and the transverse relaxation in the magnetic subsystem was neglected. The longitudinal relaxation of \mathbf{M} and \mathbf{L} has to be treated because so-called TE polaritons cannot exist in an antiferromagnet in this geometry otherwise.

Let us consider small vibrations near the equilibrium position indicated above. Linearization of the first three equations in the system (5) for the Fourier components of the variables enables us to write them in the following manner:

$$m_i = \chi_{ij}^M h_j + \chi_{ij}^{ME} e_j, \quad p_i = \kappa_{ij}^E e_j + \kappa_{ij}^{ME} h_j, \quad (6)$$

where $\hat{\chi}^{M}$ and $\hat{\varkappa}^{E}$ are the dynamic magnetic and dielectric susceptibility tensors, and $\hat{\chi}^{ME} \equiv \hat{\varkappa}^{ME}$ is the dynamic magnetoelectric susceptibility tensor. The expressions for their components in crystals with a $4_{z}^{\pm}2_{x}^{-}I^{-}$ structure have the form

$$\chi_{xx}^{M} = \chi_{yy}^{M} = \chi_{\perp} \omega_{s}^{2} (\omega_{1e}^{2} - \omega^{2}) / \Delta,$$

$$\chi_{zz}^{M} = \chi_{\parallel} \omega_{\lambda} (\omega_{2e}^{2} - \omega^{2}) / \Delta_{\lambda},$$

$$\kappa_{xx}^{E} = \kappa_{yy}^{E} = \kappa_{\perp} \omega_{1e}^{2} (\omega_{s}^{2} - \omega^{2}) / \Delta, \quad \kappa_{zz}^{E} = \kappa_{\parallel} \omega_{2e}^{2} (\omega_{\lambda} - i\omega) / \Delta_{\lambda},$$

$$\chi_{yy}^{ME} = \pm \chi_{xx}^{ME} = (\kappa_{\perp} \chi_{\perp})^{1/2} \omega_{s}^{2} \omega_{1e}^{2} \zeta_{\perp}^{1/2} / \Delta,$$

$$\chi_{zz}^{ME} = (\chi_{\parallel} \kappa_{\parallel})^{1/2} \omega_{\lambda} \omega_{2e}^{2} \zeta_{\parallel}^{1/2} / \Delta_{\lambda},$$

$$\Delta = (\omega_{s}^{2} - \omega^{2}) (\omega_{1e}^{2} - \omega^{2}) - \omega_{s}^{2} \omega_{1e}^{2} \zeta_{\perp}, \quad \zeta_{\perp} = \gamma_{3}^{2} L^{2} \chi_{\perp} \kappa_{\perp},$$

$$\omega_{\lambda} = (\omega_{\lambda} - i\omega) (\omega_{2e}^{2} - \omega^{2}) - \omega_{\lambda} \omega_{2e}^{2} \zeta_{\parallel}, \quad \zeta_{\parallel} = \gamma_{4}^{2} L^{2} \chi_{\parallel} \kappa_{\parallel},$$

$$\omega_{s}^{2} = \omega_{E} \omega_{A}, \quad \omega_{\lambda} = g L \lambda \chi_{\parallel}^{-1}, \quad \omega_{E} = g L \chi_{\perp}^{-1},$$

$$\omega_{A} = g L |\beta_{1}|, \quad \omega_{1e}^{2} = \kappa_{\perp} f, \quad \omega_{2e}^{2} = \kappa_{\parallel} f,$$

$$\chi_{\perp}^{-1} = a + D' L^{2}, \quad \chi_{\parallel}^{-1} = a + (D + D') L^{2}.$$
(7)

The remaining components of these tensors are equal to zero. In crystals with a $4_z^{\pm} 2_x^+ I^-$ structure the tensors $\hat{\chi}^M$ and $\hat{\varkappa}^E$ are also specified by Eqs. (7), but in the components of $\hat{\chi}^{ME}$ the index *yy* should be replaced by *yx* and *xx* should be replaced by *xy*. For crystals of these symmetries χ_{zz}^{ME} is equal to zero, since, according to (4), the term containing γ_4 in the magnetoelectric energy is absent. It follows from (7) that the magnetic and dielectric susceptibilities have features in the vibrational frequencies of both the magnetic and dielectric subsystems due to the magnetoelectric effect (the possibility of such a situation in solids was noted in Ref. 10). The magnetoelectric susceptibility also has features in these frequencies. The static values of these susceptibilities are expressed by the formulas

$$\chi_{xx}^{M}(0) = \chi_{\perp} (1 - \zeta_{\perp})^{-1}, \quad \kappa_{xx}^{E}(0) = \kappa_{\perp} (1 - \zeta_{\perp})^{-1},$$
$$\chi_{xx}^{ME}(0) = (\kappa_{\perp} \chi_{\perp} \zeta_{\perp})^{1/2} (1 - \zeta_{\perp})^{-1} = \gamma_{3} L \chi_{\perp} \kappa_{\perp} (1 - \zeta_{\perp})^{-1},$$
(8)

and the analogous expressions for the zz components are obtained by replacing the subscript \perp by \parallel . The magnetoelec-

tric constants γ can be evaluated from (8) using experimental values for χ^{ME} . The values of the magnetoelectric susceptibility vary over a broad range: $\chi^{ME} \sim 10^{-5} - 10^{-2}$ (Ref. 2). Hence as typical values of the parameters of the problem we obtain $L_0 \sim 10^3$ Oe, $\chi_{\perp} \sim 10^{-3}$, and $\kappa_{\perp} \sim 1$. The values of the magnetoelectric constants γ also vary in the range from 10^{-5} to 10^{-2} , and ζ varies from 10^{-7} to 10^{-1} (the latter values of γ and ζ are valid, for example, for terbium phosphate).

The presence of features at the same frequencies in $\hat{\chi}^{M}$ and $\hat{\varkappa}^{E}$, as well as the presence of a nonzero magnetoelectric susceptibility χ^{ME} , can produce new types of surface waves in magnetoelectric antiferromagnets. To obtain their dispersion laws, we must solve Maxwell's equations in the system (5) using the relations (6) and continuity at x=0 of the normal components of the magnetic induction **B** and the electric displacement **D**, and of the tangential components of the corresponding fields **H** and **E**. We seek the solution of Maxwell's equations within the antiferromagnet in the form

 $\mathbf{h}^{(i)}, \mathbf{e}^{(i)} \propto \exp(-i\omega t + ikz - \kappa_i x), \quad x > 0, \tag{9}$

and outside the antiferromagnet in the form

$$\mathbf{h}^{(e)}, \mathbf{e}^{(e)} \propto \exp(-i\omega t + ikz + \kappa_e x), \quad x < 0, \tag{10}$$

(it is assumed that the antiferromagnet has a boundary with a vacuum at x=0). Substituting (9) and (10) into Maxwell's equations for an antiferromagnet and a vacuum, we can obtain the dispersion equations of the surface waves. These equations are different for crystals of different symmetry. To illustrate the characteristic effect of the magnetoelectric interaction on the spectrum of surface waves in an antiferromagnet, we shall next consider a crystal of the $4\frac{\pm}{z}2^+_xI^-$ class, in which this influence is most pronounced in the geometry under consideration.

For the reciprocals of the penetration depths of the surface waves in the vacuum (e) and the antiferromagnetic (i), we obtain the following expressions from Maxwell's equations using (6), (7), (9), and (10) in $4_z^- 2_x^+ I^-$:

$$\kappa_e^2 = k^2 - \frac{\omega^2}{c^2},$$

$$\kappa_i^2 = \frac{1}{\mu_1} \left(k^2 + \frac{2k\omega\alpha}{c} - \frac{\omega^2\mu_1\varepsilon_1}{c^2} \right),$$
(11)
$$\kappa_i^2 = \frac{\varepsilon_2}{\varepsilon_1} \left[k^2 - \frac{2k\omega\alpha}{c} - \frac{\omega^2(\mu_1\varepsilon_1 - \alpha^2)}{c^2} \right].$$

Here $\mu_1 = 1 + 4\pi \chi_{xx}^M$, $\varepsilon_{1,2} = 1 + 4\pi \kappa_{xx,zz}^E$, and $\alpha = 4\pi \chi_{xy}^{ME}$. The second equation describes the propagation of a TE wave (h_x, h_z, e_y) within the antiferromagnet, and the first equation describes the propagation of a TM wave (e_x, e_z, h_y) . Fulfillment of the boundary conditions for the solutions (9) and (10) leads to an additional relation between k and κ , which, together with Eqs. (11), makes it possible to obtain the dispersion laws of the surface waves. For TE waves this relation has the form

$$\kappa_i = -\mu_2 \kappa_e \,, \tag{12}$$

and for TM waves it has the form

I

$$\kappa_i = -\varepsilon_2 \kappa_e \,. \tag{13}$$

Here $\mu_2 = 1 + 4\pi \chi_{zz}^M$. It is seen from (12) that surface TE polaritons are damped in the symmetry under consideration, since, according to (7), χ_{zz}^M is complex. These polaritons were considered in Ref. 11. Surface waves of another form are possible only when the component $\varepsilon_{zz} = \varepsilon_2$ of the dielectric tensor is negative. According to (7) these modes exist in the frequency range

$$\omega_{2e} \leq \omega \leq \omega_{3e} , \qquad (14)$$

where $\omega_{3e} = \omega_{2e}(1 + 4\pi\kappa_{\parallel})^{1/2} = \varepsilon_{\parallel}^{1/2}\omega_{2e}$. The range (14) lies in the optical region. According to (7), TM polaritons are nondamping, since only the longitudinal relaxation was taken into account in the original equations (5). Using (13), from (11) we ultimately obtain the dispersion equation of surface TM polaritons in magnetoelectric antiferromagnets of $4_r^2 2_x^+ I^-$ symmetry:

$$ck = \omega \frac{\alpha \pm \alpha^2 + (1 - \varepsilon_1 \varepsilon_2) [\varepsilon_1(\mu_1 - \varepsilon_2) - \alpha^2]^{1/2}}{1 - \varepsilon_1 \varepsilon_2}.$$
 (15)

Since surface TM polaritons can be observed only when $\varepsilon_2 < 0$ holds, it follows from (11) that their domain of existence is divided into three parts.

1. Polaritons of type I: $\varepsilon_2(0,\varepsilon_1)$, $\mu_1 > 0$.

In the (ω,k) plane these polaritons can exist for $\omega < \omega_{4e}$ and $\omega > \omega_{6e}$ in the region $-\omega/c_{-} < k < -\omega/c$ and in the region $\omega/c < k < \omega/c_{+}$. The values of c_{\pm} and $\omega_{4e,6e}$ are given by the formulas

$$c_{\pm} = c/(\sqrt{\mu_{1}\varepsilon_{1}} \pm \alpha),$$

$$\omega_{4e}^{2} = \omega_{1e}^{2} + \omega_{s}^{2} \omega_{1e}^{2} \zeta_{\perp} / (\omega_{1e}^{2} - \omega_{s}^{2}).$$

$$\omega_{6e}^{2} = \omega_{1e}^{2} \varepsilon_{\perp} + \omega_{s}^{2} \omega_{1e}^{2} \zeta_{\perp} / (\omega_{1e}^{2} \varepsilon_{\perp} - \omega_{s}^{2}), \quad \varepsilon_{\perp} = 1 + 4\pi\kappa_{\perp}.$$
(16)

The frequencies ω_{4e} and ω_{6e} are the optical roots of the equations $\Delta=0$ and $\varepsilon_1=0$, respectively. In the derivation of the expressions for ω_{4e} and ω_{6e} it was assumed that the condition $\omega_s^2 \ll \omega_{1e}^2$ is satisfied.

2. Polaritons of type II: $\varepsilon_2 < 0$, $\varepsilon_1 < 0$, $\mu_1 < 0$.

Polaritons of this type can be observed for $\omega_{4e} < \omega < \omega_{5e}$ and in the regions $k > \max(\omega/c_+, \omega/c)$ and $k < \min(-\omega/c_-, -\omega/c)$. The frequency

$$\omega_{5e} = \sqrt{\omega_{1e}^2 + \omega_s^2 \omega_{1e}^2 \zeta_{\perp} / (\omega_{1e}^2 - \mu_{\perp} \omega_s^2)}$$
(1)

is the optical root of the equation $\mu_1=0$.

3. Polaritons of type III: $\varepsilon_2 < 0$, $\varepsilon_1 < 0$, $\mu_1 > 0$.

These polaritons can exist for $\omega_{5e} < \omega < \omega_{6e}$ and any k in the ranges $\omega < -ck$ and $\omega < ck$.

We note that all the surface polaritons just enumerated will be observed only when their domains of existence fall in the range (14). We also note that the polaritons of type I are virtual (relativistic): when there is no retardation $(c \rightarrow \infty)$, they are absent. The polaritons of types II and III can be real, if the frequency

$$\omega_{7e} \approx \sqrt{\omega_{1e}^2(\varepsilon_{\perp} + \beta)/(1 + \beta^2) + \omega_s^2 \zeta_{\perp}/(\varepsilon_{\perp} + \beta)}$$
(18)

 $(\beta = \kappa_{\perp} / \kappa_{\parallel})$, which is the optical root of the equation $1 - \varepsilon_1 \varepsilon_2 = 0$, falls in their domain of existence and in the frequency range (14). At this frequency the wave number (15) tends to infinity in the region of positive k.

FIG. 1. Sign-reversal scheme of the magnetic permeability (μ_1) and the dielectric constant (ε_1) as a function of the frequency. The frequencies ω_{4e} to ω_{6e} are the optical roots of the equations $\Delta=0$, $\mu_1=0$, and $\varepsilon_1=0$, respectively. Here $\alpha > 0$ when $\omega < \omega_{4e}$. The Roman numerals label the possible regions for the existence of surface modes of different types, if the range (14), where $\varepsilon_2 < 0$, falls in these regions.

Thus, it can be concluded that new types of surface waves are possible in magnetoelectric antiferromagnets. They include, in particular, surface modes of type II. These excitations arise because the magnetic permeability can be negative even at optical frequencies in antiferromagnets when the magnetoelectric effect is taken into account. We note that the character of surface modes of type I and III in magnetoelectric interaction, the spectrum of these modes is asymmetric relative to the frequency axis, i.e., nonreciprocity appears, which is confined to the fact that the velocities of the surface waves along the z axis in the positive and negative directions are not equal to one another. The velocity difference is determined by the magnitude of the magnetoelectric coupling.

The specific form of the spectrum of surface TM polaritons depends on the relationship between κ_{\perp} and κ_{\parallel} . Figure 1 presents the sign-reversal scheme of the magnetic permeability μ_1 and the dielectric constant ε_1 as a function of the frequency in the optical range. The frequency range (14) for the existence of surface TM polaritons can be found in any of the regions indicated in Fig. 1 in the general case. Its location depends on the relationship between the values of the parameters $\beta = \kappa_{\perp}/\kappa_{\parallel}$, $\varepsilon_{\parallel} = 1 + 4\pi\kappa_{\parallel}$, and ε_{\perp} . An analysis reveals that only four cases are possible:

1.
$$\beta \geq \varepsilon_{\parallel}, \quad \omega_{6e} \geq \omega_{5e} \geq \omega_{4e} \geq \omega_{3e} \geq \omega_{2e}, \quad (19)$$

polaritons of type I;

7)

2.
$$1 \leq \beta < \varepsilon_{\parallel}$$
, $\omega_{6e} > \omega_{3e} > \omega_{5e} > \omega_{4e} > \omega_{2e}$,

polaritons of types I, II, and III;

3.
$$1/\varepsilon_{\perp} \leq \beta < 1$$
, $\omega_{3e} > \omega_{6e} > \omega_{2e} > \omega_{5e} > \omega_{4e}$

polaritons of types I and III;

4. $\beta < 1/\varepsilon_{\perp}$, $\omega_{3e} > \omega_{2e} > \omega_{6e} > \omega_{5e} > \omega_{4e}$,

polaritons of type I.

When $\beta > \varepsilon_{\parallel}$ holds, we have $\varepsilon_2 \rightarrow -\infty$ on the lower boundary of the range (14), and, according to (15), the wave number is $k = \pm \omega_{2e}/c$. On the upper boundary of the range we have $\varepsilon_2 \rightarrow 0$, and the wave number is determined by the formula $k = \pm \omega_{3e}/c_{\pm}(\omega_{3e})$. Thus, the domain of existence of polaritons of type I is confined to the region between the straight lines $\omega = \pm ck$ and $\omega = \pm c_{\pm}(\omega_{3e})k$. The dispersion curves of the polaritons of type I for the first case are pre-



FIG. 2. Dispersion curves of polaritons of type I when $\beta > \varepsilon_{\parallel}$. The dashed lines are the curves for $\omega_{3e} \rightarrow \omega_{4e}$ ($\beta = \varepsilon_{\parallel}$).

sented in Fig. 2. It is seen that these polaritons are, in fact, virtual, since they are absent when $c \rightarrow \infty$ (i.e., in the abscence of retardation). Only when we have $\beta = \varepsilon_{\parallel}$ (or $\omega_{3e} = \omega_{4e}$), do these polaritons transform into real polaritons in the region of positive k (the dashed curves in Fig. 2). When $\omega = \omega_{2e}$ holds, the penetration depth κ_i^{-1} of the surface waves in the antiferromagnetic is equal to zero, and in the vacuum κ_e^{-1} is equal to infinity. On the upper boundary of the domain of existence of polaritons of type I, the penetration depth κ_i^{-1} equals infinity, and in the vacuum it is given by the formula

$$\kappa_e^{-1} = \omega_{3e} [(\sqrt{\mu_1 \varepsilon_1} \pm \alpha)^2 - 1]^{1/2} / c, \qquad (20)$$

where the plus and minus signs correspond to positive and negative k. It follows from Fig. 2 that the spectrum of polaritons is asymmetric relative to the frequency axis. This indicates that the waves propagate at different speeds in the positive and negative directions along the z axis. The difference between these speeds is determined by the strength of the magnetoelectric coupling. This nonreciprocity arises because the magnetoelectric susceptibility tensor is gyrotropic.

Surface polaritons of all three types can appear in the range $\omega_{2e} < \omega < \omega_{3e}$ for $1 < \beta < \epsilon_{\parallel}$ (19). For $\omega_{3e} < \omega < \omega_{4e}$ the surface modes are polaritons of type I, for $\omega_{4e} < \omega < \omega_{5e}$ they are polaritons of type II, and for $\omega_{5e} < \omega < \omega_{7e}$ they are polaritons of type III. The dispersion dependence of the polaritons (Fig. 3) is practically continuous, although the char-



FIG. 3. Dispersion dependence of the surface polaritons for $1 \le \beta \le \varepsilon_{\parallel}$. The Roman numerals label the domains of the existence of polaritons of different types.



FIG. 4. Dispersion curve of surface polaritons of type III for $1/\epsilon_{\perp} < \beta < 1$.

acter of the modes changes at $\omega = \omega_{4e}$ and $\omega = \omega_{5e}$, where modes of one type are replaced by other modes. At the lower boundary of the range (14) the value of the wave number is $k = \pm \omega_{2e}/c$. The penetration depths of the waves in the antiferromagnet and the vacuum are equal to zero and infinity, respectively. When $\omega \rightarrow \omega_{7e}$ (18), the polaritons of type III transform into real polaritons $(k \rightarrow \infty \text{ when } \omega \rightarrow \omega_{7e})$ in the positive range of wave numbers, and the surface polaritons are virtual over the entire frequency domains of existence when k is negative. At the upper boundary of the domain of existence of the surface modes $(\omega = \omega_{7e})$ the penetration depth of the polaritons tends to zero both in the antiferromagnet and in the vacuum in the region of positive k, and the surface modes convert into bulk modes occurs (κ_i and κ_e become imaginary) in the region of negative k.

According to (19), polaritons of types I and III should exist in the range (14) when $1/\varepsilon_{\perp} < \beta < 1$. However, a more detailed analysis reveals that only polaritons of type III can appear in this case. The dispersion curve is presented in Fig. 4. The values of the wave numbers and the behavior of the penetration depth in the antiferromagnet and the vacuum at the lower boundary of the range (14) are the same as in the two preceding cases. At the upper boundary the behavior of these parameters is similar to their behavior in the preceding case. The wave number k_1 is given by the formula (as in Fig. 3)

$$k_1 = -\frac{\omega_{7e}}{c} \frac{\varepsilon_1(\mu_1 - \varepsilon_2) - \alpha^2}{2\alpha}, \qquad (21)$$

where the values of ε_1 , μ_1 , ε_2 , and α should be calculated for $\omega = \omega_{7e}$. Hence it is seen that k_1 depends strongly on the magnetoelectric coupling constant γ_3 , which appears in the expression (11) for α . When $\gamma_3 \rightarrow 0$, the surface modes of type III in the region of negative k also transform into real polaritons.

In the last case, in which $0 < \beta < 1/\varepsilon_1$, only polaritons of type I should appear according to (19). However, here, too, a more detailed analysis of (11) and (15) reveals that these polaritons cannot exist in the range (14).

Thus, magnetoelectric antiferromagnets, unlike ordinary antiferromagnets, should exhibit the following main features. Consideration of the magnetoelectric coupling results, first, in asymmetry of the dispersion curves relative to the frequency axis. This corresponds to the appearance of nonreci-

TABLE I. Components of the magnetoelectric tensor. DIT 2) II 2⁺/⁻ 3) II 27/ 4) III $2^+_{x}2^+_{y}I^{-1}$ 6) III $2^{-}_{x}2^{+}_{x}I^{-}$ 5) III $2^+_x 2^-_y l^-$ 7) III 2, 2, 1 10) IV $4_z^+ 2_d^+$ 12) IV $4_z^2 2_d^+ I^2$ 11) IV $4_z^+ 2_d^- l^ (\cdots, \cdots, \cdots, \cdots)$ 13) IV 4,2,1 14) V 3⁺/ 15) V 3, 2, 1 (· · · · ·) 16) V 3,2,1 17) VI 6,⁺/ 18) VI 67 $(\cdots, \cdots, \cdots)_{\alpha}$ $(\cdots, \cdots, \cdots)_{\alpha}$ $(\cdots, \cdots, \cdots)_{\alpha}$ $(\underbrace{0}, \underbrace{0}, \underbrace{0$ 22) VI $6_{z}^{-} 2_{z}^{-} / \overline{}^{-}$ 23) VII $2_{z}^{+} 3_{1111}^{+} / \overline{}^{-}$ 24) VII $4_{z}^{+} 3_{1111}^{+} / \overline{}^{-}$ 25) VIII $4^{-3^{+}}_{2^{-1}}$

procity, i.e., a difference between the rates of propagation of the surface waves along the antiferromagnetism vector and in the opposite direction. Second, at negative wave numbers the surface modes become virtual. When $c \rightarrow \infty$, these modes are absent. Special attention must be focused on this fact in experimental investigations of these modes. Third, surface modes of a new type can appear in magnetoelectric antiferromagnets. They are polaritons of type II. Their appearance is due to the dispersion of the magnetic permeability μ_1 in the optical range. This dispersion, in turn, results specifically from the magnetoelectric coupling. The polaritons of type II are magnetic in nature; therefore, they should be called "optical magnetic" (or "magneto-optical") polaritons.

We note that in the present work we restricted ourselves to a treatment of one class of tetragonal magnetoelectric antiferromagnets, viz., $4_z^- 2_x^+ I^-$, and one geometry of the problem. An investigation of other classes of antiferromagnets and other geometries would significantly expand the scope of the present work. Therefore, we shall indicate here only the main phenomena appearing in other problems.

For antiferromagnets of the $4_z^+ 2_x^+ I^-$ class and the ge-

ometry indicated above, the sign in front of α should be reversed, according to (7) and (11). This is equivalent to reversing the sign in front of the wave number in (11) and (15) while maintaining the sign of α . Thus, the spectrum of the surface polaritons in $4_z^+ 2_x^+ I^-$ crystals in the geometry considered can be obtained from the spectrum presented here via the replacement $k \rightarrow -k$, i.e., upon mirror reflection of the spectrum investigated relative to the frequency axis.

In magnetoelectric antiferromagnets of the $4_z^{\pm} 2_x^* I^$ classes in a geometry in which the antiferromagnetism vector is perpendicular to the surface of the sample and the wave vector of the surface wave, both TE polaritons and TM polaritons can appear. One special feature of these polaritons is that they can appear both at vibrational frequencies of the spin subsystem ($\omega \sim \omega_s$) and at optical frequencies ($\omega \sim \omega_{1e}$). Another important feature in such a geometry is that all the types of surface polaritons are modes which appear in the bulk of the antiferromagnet.

In antiferromagnets of the $4\frac{t}{z}2_x^-I^-$ classes in the geometry considered here, the models of the TE and TM polaritons are coupled. In addition, the presence of the term

 $-\gamma_4 M_z P_z L_z$ causes the component μ_{zz} of the magnetic permeability tensor to also have a feature in the optical frequencies. Thus, the spectrum of surface polaritons in antiferromagnets of the $4_z^{\pm} 2_x^{-} I^{-}$ classes should differ considerably from the spectrum considered above. In particular, weakly damped TE surface polaritons should be expected to occur in such magnets. An analysis of the spectrum of the surface modes in tetragonal antiferromagnets of the $4_z^{\pm} 2_x^{-} I^{-}$ classes requires a separate investigation.

The substances in which the surface polaritons considered here might be observed should be mentioned. They include, for example, trirutiles¹²⁻¹⁴ and rare-earth phosphates and vanadates,^{1,15,16} if the ground state considered here is realized in them. In particular, the highest magnetoelectric susceptibility $\chi^{ME} \sim 10^{-2}$ was discovered experimentally in tetragonal terbium phosphate.¹

Finally, we recall that the spatial dispersion of dielectric constant was neglected in the present work. In particular, this can have bearing on the fact that the vanishing of the penetration depth of the surface waves in the magnet renders the macroscopic approach considered here inapplicable. The usual condition for macroscopic waves is $ak \ll 1$, where a is the lattice constant. It was shown in Ref. 17 that consideration of the spatial dispersion of the magnetic permeability tensor (2) leads to a more rigid condition than the usual condition for macroscopic waves. Thus, all the results obtained here hold when $ak \ll 1$.

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APPENDIX A

The table presents the nonzero components of the magnetoelectric coefficient tensor $\hat{\gamma}$ in the energy F_{ME} $= -\gamma_{\alpha\beta\gamma}M_{\alpha}L_{\beta}P_{\gamma}$ for crystals of different symmetry. The filled circles denote nonzero components of $\hat{\gamma}$, and the unfilled circles denote nonzero components which must be written with a minus sign. The lines connect components which are equal in absolute value. The number of independent components of $\hat{\gamma}$ is given in parentheses. For simplicity the tensor is written in a two-index scheme: $\gamma_{\alpha\beta\gamma} = \gamma_{\alpha j}$, where the values j=1, 2, 3, 4, 5, 6, 7, 8, and 9 correspond to the indices $\beta\gamma = 11, 12, 13, 21, 22, 23, 31, 32, and 33$.

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