PHYSICAL REVIEW E **93**, 052603 (2016)

Shear modulus of structured electrorheological fluid mixtures

Kyohei Shitara* and Takahiro Sakaue†

Department of Physics, Kyushu University, 744 Motooka, Nishi-ku, Fukuoka 819-0395, Japan

(Received 26 November 2015; revised manuscript received 24 March 2016; published 5 May 2016)

Some immiscible blends under a strong electric field often exhibit periodic structures, bridging the gap between two electrodes. Upon shear, the structures tilt, and exhibit an elastic response which is mostly governed by the electric energy. Assuming a two-dimensional stripe structure, we calculate the Maxwell stress, and derive an expression for the shear modulus, demonstrating how it depends on the external electric field, the composition, and the dielectric properties of the blend. We also suggest the notion of effective interfacial tension, which renormalizes the effect of the electric field. This leads to a simple derivation of the scaling law for the selection of the wavelength of the structure formed under an electric field.

DOI: 10.1103/PhysRevE.93.052603

I. INTRODUCTION

When applying an electric field to some specific fluids, their apparent viscosity increases sharply [1]. The fluids exhibiting such an effect are called electrorheological fluids (ER fluids). There are movements to apply ER fluids to some products, such as a brake and clutch system [2,3]. Ordinary ER fluids are suspensions which consist of insulating liquids such as a silicon oil and nonconducting small particles and exhibit a high apparent viscosity by the following mechanism [4]: Once an electric field is applied to the suspensions, electric dipoles appear in the particles. Dipoles aligned with the electric field attract each other, while dipoles with their lines of center normal to the electric field repel each other. Because of the interactions, the particles aggregate into fibrous columns perpendicular to the electrodes and construct structures spanning the electrode gap. This dramatic change in the structures increases the apparent viscosity of the suspensions. There are some disadvantages of ER suspensions, however, related to a degradation of the ER effect due to aggregation, sedimentation, solidification of the particles, the wear and tear between the particles and the electrodes, etc. [5]. Different types of ER fluids have been developed to solve these kinds of problems [6–9]. Binary mixture fluids such as polymer blends are one of the candidates [10–16]. A mixture of dielectric fluids 1 and 2, which are immiscible, exhibits an electrorheological response when their dielectric constants $\varepsilon_1 = \bar{\varepsilon} + \delta\varepsilon$ and $\varepsilon_2 = \bar{\varepsilon} - \delta\varepsilon$ are different. Here, the electrorheological response arises from Maxwell stress acting on the interface.

Recently, we have suggested that the effect of Maxwell stress on the average stress can be effectively treated as anisotropic interfacial tension [17]. In Ref. [18], we then proposed a set of constitutive equations describing the electrorheological behavior of immiscible binary fluid mixtures when the effect of the electric field is relatively weak. However, the notion of anisotropic interfacial tension due to Maxwell stress would be most useful under a strong electric field. In fact, the condition of a weak electric field is expressed as $\mathcal{S} \ll 1$, with the dimensionless number

$$\mathcal{S} \equiv \frac{K_\varepsilon E_{\rm ex}^2 \lambda}{\Gamma}, \tag{1}$$

where $K_\varepsilon = (\delta\varepsilon)^2/\bar{\varepsilon}$, $E_{\rm ex}$ is the external electric field (defined as the voltage difference across the capacitor divided by its gap width), Γ is the interfacial tension, and λ is the typical domain size in the mixture. In such a situation, $\mathcal{S} \ll 1$, the electric field weakly perturbs the domain morphology. Note that under a weak electric field, the domain size in steady shear flow (with shear rate $\dot{\gamma}$) is determined by the balance between the interfacial and viscous stresses, i.e., $\lambda \simeq \Gamma/\eta_0\dot{\gamma}$, where η_0 is the viscosity of fluids 1 and 2, which is assumed to be the same, for simplicity. Therefore, Eq. (1) becomes $\mathcal{S} = K_\varepsilon E_{\rm ex}^2/(\eta_0\dot{\gamma})$.

In the present paper, we shall look at the other limit of the strong electric field, $\mathcal{S} \gg 1$. Here, the mixture typically forms a columnar structure, which has a resistance to shear deformation, i.e., the mixture exhibits a solidlike response. Experimentally, a plateau shear modulus is observed over a wide frequency range [19]. By considering a two-dimensional (2D) stripe geometry, we derive several important features of the binary fluid mixture under a strong electric field, including the wavelength of the stripe and the plateau shear modulus. In Sec. III, we first calculate the Maxwell stress of the system based on the perturbation theory. In Sec. IV, we perform a standard electrostatic analysis, which provides an alternative derivation of the Maxwell stress. In Sec. V, we then argue that the Maxwell stress can be expressed using an interfacial tensor, which leads to the notion of renormalized effective interfacial tension. This allows us to evaluate the typical wave number of the stripe structure. The analysis of the 2D stripe is mostly motivated by its simple tractability, but we expect that the essences of the obtained results apply to the 3D case as well, as briefly discussed in the last part of Sec. III.

II. ELASTIC STRESS IN DIELECTRIC FLUIDS UNDER STRONG ELECTRIC FIELD

We consider a binary fluid which consists of components 1 and 2, and introduce the order parameter $\phi(\vec{r}) = \phi_1(\vec{r}) - \phi_2(\vec{r})$, where $\phi_1(\vec{r})$ and $\phi_2(\vec{r})$ are local volume fractions of components 1 and 2, respectively. Note that the order parameter $\phi(\vec{r})$ is normalized to lie between -1 and 1, and the condition $\phi_1(\vec{r}) + \phi_2(\vec{r}) = 1$ is always satisfied. We consider the sharp

*Present address: Center for Simulation Sciences, Ochanomizu University, 2-1-1 Otsuka, Bunkyo-ku, Tokyo 112-8610, Japan; shitara.kyohei@ocha.ac.jp

†sakaue@phys.kyushu-u.ac.jp

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interface limit, where the phase-separated domain structure is characterized by the interfacial tensor (see below). The elastic shear stress of the mixture can be decomposed into the interfacial stress $\sigma^I_{\alpha\beta}$ and the Maxwell stress $\sigma^M_{\alpha\beta}$.

Interfacial stress. The average stress arising from the interfacial energy depends on the configuration of the interface. Introducing an interfacial tensor [20]

$$q_{\alpha\beta} = \frac{1}{\Omega}\int dS(n_\alpha n_\beta), \tag{2}$$

where $\int dS()$ is the surface integral, Ω is a volume of the system, and $\vec{n}$ is the unit vector normal to the interface, the interfacial stress $\sigma^I_{\alpha\beta}$ is given as

$$\sigma^I_{\alpha\beta} = -\Gamma q_{\alpha\beta}. \tag{3}$$

Maxwell stress. The local Maxwell stress $\sigma^M_{\alpha\beta}(\vec{r}) = \varepsilon(\vec{r})E_\alpha(\vec{r})E_\beta(\vec{r})$ depends on the dielectric constant and the electric field at the local position, which depend on the domain structure of the mixture in a nontrivial way. There is a useful formula to calculate the average Maxwell stress $\sigma^M_{\alpha\beta} = \Omega^{-1}\int d\vec{r}\,\sigma^M_{\alpha\beta}(\vec{r})$ in terms of the structure factor $\langle\delta\phi_{\vec{k}}\,\delta\phi_{-\vec{k}}\rangle$ of the system [17,21]:

$$\sigma^M_{\alpha\beta} = -\frac{K_\varepsilon}{\Omega}\int_{\vec{k}}\frac{(k_\alpha E_{\mathrm{ex},\beta} + k_\beta E_{\mathrm{ex},\alpha})(\vec{k}\cdot\vec{E}_{\mathrm{ex}})}{k^2}\langle\delta\phi_{\vec{k}}\,\delta\phi_{-\vec{k}}\rangle + \frac{K_\varepsilon}{\Omega}\int_{\vec{k}}\frac{k_\alpha k_\beta(\vec{k}\cdot\vec{E}_{\mathrm{ex}})^2}{k^4}\langle\delta\phi_{\vec{k}}\,\delta\phi_{-\vec{k}}\rangle. \tag{4}$$

Here, $\phi_{\vec{k}} = \int d\vec{r}\delta\phi(\vec{r})e^{i\vec{k}\cdot\vec{r}}$ with $\delta\phi(\vec{r}) = \phi(\vec{r}) - \phi_0$, where ϕ_0 is a spatial average of $\phi(\vec{r})$ defined as $\phi_0 = \Omega^{-1}\int d\vec{r}\phi(\vec{r})$. This formula is based on the perturbative calculation of the local electric field, and would be valid for $\delta\varepsilon/\bar{\varepsilon} \ll 1$. As already stated, $\vec{E}_{\mathrm{ex}}$ is the external electric field, which is constant in space, and accordingly all the spatial structural information is contained in the structure factor $\langle\delta\phi_{\vec{k}}\,\delta\phi_{-\vec{k}}\rangle$ in this formula.

In the present situation, the sample with the volume $\Omega = Sl$ is confined between two parallel plates with area S with their separation l, and a constant voltage difference V is applied between the plates. We take the normal to the plate direction as the y axis, and shear the sample to the x direction. Then, $\vec{E}_{\mathrm{ex}} = {}^t(0,V/l,0)$, and we define $E_{\mathrm{ex}} \equiv |\vec{E}_{\mathrm{ex}}| = V/l$. The superscript t represents a transpose sign. In this case, the above formula becomes

$$\sigma^M_{xy} = -\frac{K_\varepsilon}{\Omega}E^2_{\mathrm{ex}}\int_{\vec{k}}\left[\frac{k_xk_y}{k^2} - \frac{k_xk_y^3}{k^4}\right]\langle\delta\phi_{\vec{k}}\,\delta\phi_{-\vec{k}}\rangle. \tag{5}$$

III. PERTURBATION CALCULATION OF MAXWELL STRESS

Let us consider the 2D system, where the fluid mixture forms a stripe structure with the spatial period λ under a strong electric field. The mixture is confined between parallel plates with a separation l and a surface area (length in 2D) S. The voltage drop V across the plate is constant. The geometry of the system is shown in Fig. 1.

When small shear deformation with shear strain γ is imposed on the system, $\delta\phi(\vec{r})$, the spatial modulation from

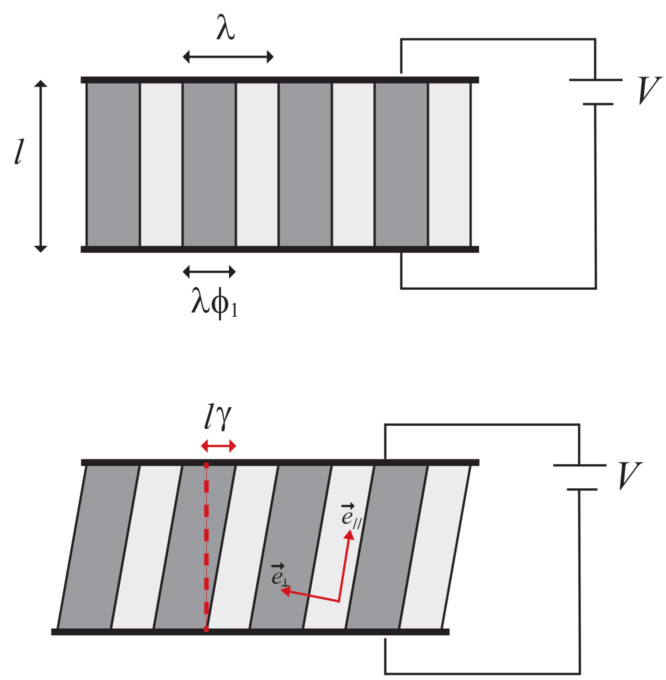

FIG. 1. Geometry of the system. Upper: No shear strain. Lower: Weak shear strain γ is applied.

ϕ_0 is represented as

$$\delta\phi(\vec{r}) = 2\sum_{n=-\frac{N}{2}}^{\frac{N}{2}} H\left(x - \gamma y - n\lambda + \frac{a}{2}\right) \times H\left(\gamma y + n\lambda + \frac{a}{2} - x\right) - 1 - \phi_0, \tag{6}$$

where $H(x)$ is the Heaviside step function, $a = \lambda\phi_1$ represents a width of a region with a high value of the order parameter, ϕ_1 is a spatial average of $\phi_1(\vec{r})$, and ϕ_2 is defined in the same way as ϕ_1. We consider that the number of stripes N is large, i.e., $N \gg 1$. Due to its periodicity, $\delta\phi(\vec{r})$ can be expanded to the Fourier series,

$$\delta\phi(\vec{r}) = \sum_{m,n} C_{m,n}\exp\left(\frac{2\pi imx}{\lambda}\right)\exp\left(\frac{2\pi in\gamma y}{\lambda}\right), \tag{7}$$

where the Fourier coefficients are

$$C_{m,n} = \frac{\gamma}{\lambda^2}\int_0^\lambda\int_0^{\lambda/\gamma}\delta\phi(x,y)\exp\left(-\frac{2\pi i(mx + n\gamma y)}{\lambda}\right)dxdy. \tag{8}$$

With $\delta\phi(\vec{r})$ given by Eq. (6), one finds $C_{m,n}$ is nonzero only if $m + n = 0$. Thus, $C_{m,n}$ can be represented as C_n and

$$C_n = \begin{cases}\frac{2}{n\pi}\sin\left(\frac{n\pi a}{\lambda}\right) & (n \neq 0),\\ 0 & (n = 0).\end{cases} \tag{9}$$

Hence, Eq. (7) becomes

$$\delta\phi(\vec{r}) = \sum_{n\neq 0}\frac{2}{n\pi}\sin\left(\frac{n\pi a}{\lambda}\right)\exp\left(\frac{2n\pi i}{\lambda}(x - \gamma y)\right), \tag{10}$$

and the Fourier transformation of $\delta\phi(\vec{r})$ is

$$\delta\phi_{\vec{k}} = 8\pi^2 \sum_{n\neq 0} \frac{1}{n\pi} \sin\left(\frac{n\pi a}{\lambda}\right)\delta\left(k_x + \frac{2n\pi}{\lambda}\right) \times \delta\left(k_y - \frac{2n\pi\gamma}{\lambda}\right), \tag{11}$$

where $\delta(x)$ is the Dirac delta function. Therefore, $\langle\delta\phi_{\vec{k}}\,\delta\phi_{-\vec{k}}\rangle$ in Eq. (5) is given as

$$\langle\delta\phi_{\vec{k}}\,\delta\phi_{-\vec{k}}\rangle = 64\pi^4 \sum_{n\neq 0} \frac{1}{n^2\pi^2} \sin^2\left(\frac{n\pi a}{\lambda}\right)\delta^2\left(k_x + \frac{2n\pi}{\lambda}\right) \times \delta^2\left(k_y - \frac{2n\pi\gamma}{\lambda}\right). \tag{12}$$

By rewriting the square of the delta function as

$$\delta^2(k) = \delta(k)\frac{1}{2\pi}\lim_{L\to\infty}\int_{-L/2}^{L/2} e^{ikx} = \delta(k)\frac{1}{2\pi}\lim_{L\to\infty} L, \tag{13}$$

with a linear system size L, that is, S (l) in the x (y) direction, we obtain

$$\langle\delta\phi_{\vec{k}}\delta\phi_{-\vec{k}}\rangle = 16\pi^2\Omega \sum_{n\neq 0} \frac{1}{n^2\pi^2} \sin^2\left(\frac{n\pi a}{\lambda}\right)\delta\left(k_x + \frac{2n\pi}{\lambda}\right) \times \delta\left(k_y - \frac{2n\pi\gamma}{\lambda}\right). \tag{14}$$

We substitute Eq. (14) into Eq. (5) and write the Maxwell stress as

$$\sigma_{xy}^M \simeq B_2 K_\varepsilon E_{\rm ex}^2 \gamma, \tag{15}$$

with a factor

$$B_2 = 4\sum_{n\neq 0} \frac{1}{\pi^2 n^2} \sin^2(\pi n \phi_1), \tag{16}$$

up to the first order of a shear strain γ. For a symmetrical blend $\phi_1 = 1/2$, one can evaluate the factor as

$$B_2 = 4\sum_{n\neq 0} \frac{1}{2\pi^2 n^2}[1 + (-1)^{n+1}] = 1, \tag{17}$$

with the following formula,

$$\sum_{n=1}^{\infty} \frac{1}{n^2} = \frac{\pi^2}{6}. \tag{18}$$

For general ϕ_1, we numerically evaluate the factor B_2. The result shown in Fig. 2 indicates the maximum stress at the symmetrical composition.

In 3D systems, the mixtures often form a hexagonal structure under a strong electric field [22]. In such a case, too, it is rather straightforward to see that the scaling structure of the Maxwell stress given by Eq. (15) is preserved. Now, before shear deformation, the columns are aligned to the y direction, and the structure factor can be expressed as

$$\langle\delta\phi_{\vec{k}}\,\delta\phi_{-\vec{k}}\rangle = S_\perp(k_x,k_z)\delta(k_y), \tag{19}$$

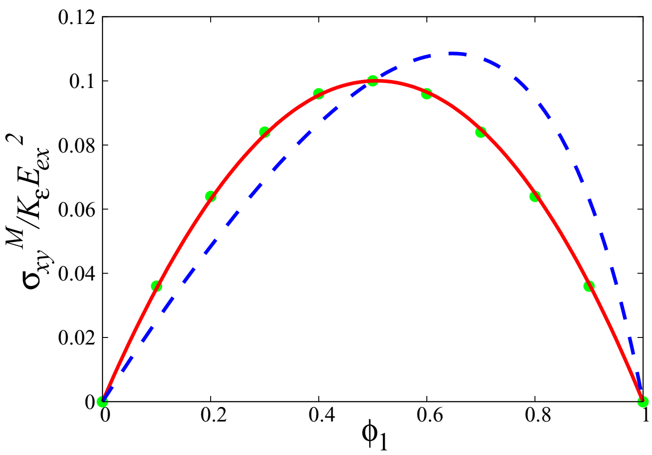

FIG. 2. The dependence of the Maxwell stress rescaled by $K_\varepsilon E_{\rm ex}^2$ on volume fraction ϕ_1. The dots are results from Eq. (15), where we have carried out the summation in Eq. (16) from $n = 1$ to $n = \pm 10^5$, and the solid line is Eq. (31) with Eq. (28). In both cases, the parameters are chosen as follows: $\varepsilon_1 = 10.0$, $\varepsilon_2 = 9.5$, $\gamma = 0.1$. The dashed line indicates Eq. (31) with Eq. (28) for the case with a relatively large dielectric constant difference ($\varepsilon_1 = 10.0$, $\varepsilon_2 = 3.0$, with other parameters being the same as before), where the deviation from the perturbation calculation (maximum stress at $\phi_1 = 1/2$) becomes evident.

where $S_\perp$ reflects the in-plane (x-z plane) structure, which is assumed to have a characteristic length scale λ. Considering that the integral is dominated by the contribution from the first peak in $S_\perp$, we approximate the structure factor as

$$\langle\delta\phi_{\vec{k}}\,\delta\phi_{-\vec{k}}\rangle \simeq \Omega\frac{\delta(k_\perp - k_\lambda)}{k_\perp} S_\theta(\theta)\delta(k_y), \tag{20}$$

where $k_\perp = (k_x^2 + k_z^2)^{1/2}$, $(k_x,k_z) = k_\perp(\cos\theta, \sin\theta)$, and $k_\lambda = 2\pi/\lambda$. When sheared, it becomes

$$\langle\delta\phi_{\vec{k}}\,\delta\phi_{-\vec{k}}\rangle \simeq \Omega\frac{\delta(k_\perp - k_\lambda)}{k_\perp} S_\theta(\theta)\delta(k_y + \gamma k_x). \tag{21}$$

Plugging this into Eq. (5), one can show the second term is negligibly small, and find

$$\sigma_{xy}^M \simeq B_3 K_\varepsilon E_{\rm ex}^2 \gamma + O(\gamma^3), \tag{22}$$

where the numerical constant

$$B_3 = \int d\theta \cos^2\theta\, S_\theta(\theta) \tag{23}$$

reflects the in-plane structure of the mixture.

IV. ELECTROSTATIC ENERGY OF DIELECTRIC FLUID CAPACITOR

The preceding calculation of the Maxwell stress is based on the perturbation calculation of the electric field, the result of which is thus valid only for the small $\delta\varepsilon/\bar{\varepsilon}$ case. Now we provide an alternative derivation of the Maxwell stress which is not limited to the small $\delta\varepsilon/\bar{\varepsilon}$ case. Again, we assume a 2D stripe geometry (Fig. 1). The electrostatic energy of the system is

$$\begin{aligned} U_E &= -\frac{1}{2}\int d\vec{r}\,\vec{E}(\vec{r})\vec{D}(\vec{r}) \\ &= -\frac{1}{2}\int d\vec{r}\varepsilon(\vec{r})[E_\parallel^2(\vec{r})+E_\perp^2(\vec{r})] \\ &= -\frac{1}{2}Sl\left[\phi_1\left(\varepsilon_1\bar{E}_\parallel^2+\frac{1}{\varepsilon_1}\bar{D}_\perp^2\right)+\phi_2\left(\varepsilon_2\bar{E}_\parallel^2+\frac{1}{\varepsilon_2}\bar{D}_\perp^2\right)\right], \end{aligned} \tag{24}$$

where we decompose the electric field vector into the parallel and the perpendicular components $\vec{E}(\vec{r})=(E_\parallel(\vec{r}),E_\perp(\vec{r}))$ to the tilted stripe direction. In the last expression, we introduce the spatial averages of $E_\parallel$ and $D_\perp$, which are marked by the bar.

To obtain $\bar{E}_\parallel$, we evaluate its line integral along the parallel path to the tilted stripe (see Fig. 1). With the path length as $l_\parallel = l(1+\gamma^2)^{1/2}\simeq l(1+\gamma^2/2)$, we have the relation

$$\bar{E}_\parallel l_\parallel = V \Leftrightarrow \bar{E}_\parallel \simeq E_{\rm ex}\left(1-\tfrac{1}{2}\gamma^2\right). \tag{25}$$

Similarly, $\bar{D}_\perp$ can be obtained by its line integral along the perpendicular path to the stripe (see Fig. 1), whose length is $l_\perp = l(1+\gamma^{-2})^{1/2}\simeq l/\gamma$. Noting that a fraction $l_\perp\phi_1$ of the total path length belongs to the domain of the component 1, we find

$$\bar{E}_{\perp,1}l_\perp\phi_1+\bar{E}_{\perp,2}l_\perp\phi_2 = V \Leftrightarrow \bar{D}_\perp \simeq E_{\rm ex}\gamma\frac{\varepsilon_1\varepsilon_2}{\varepsilon_2\phi_1+\varepsilon_1\phi_2}. \tag{26}$$

Plugging Eqs. (25) and (26) into Eq. (24), we obtain the following expression for the electrostatic energy of the system subjected to a small strain $\gamma \ll 1$:

$$U_E \simeq -\tfrac{1}{2}SlE_{\rm ex}^2[\varepsilon_1\phi_1+\varepsilon_2\phi_2-\gamma^2 g(\varepsilon_1,\varepsilon_2,\phi_1)], \tag{27}$$

with a function

$$\begin{aligned} g(\varepsilon_1,\varepsilon_2,\phi_1) &\equiv -\frac{\varepsilon_1\varepsilon_2}{\varepsilon_2\phi_1+\varepsilon_1\phi_2}+\varepsilon_1\phi_1+\varepsilon_2\phi_2 \\ &= -\frac{\varepsilon_1\varepsilon_2}{\varepsilon_1+(\varepsilon_2-\varepsilon_1)\phi_1}+\varepsilon_2-(\varepsilon_2-\varepsilon_1)\phi_1. \end{aligned} \tag{28}$$

In the following, we just describe $g(\varepsilon_1,\varepsilon_2,\phi_1)$ as g.

The Maxwell stress can be derived from the principle of virtual work as in the following way. Let δU_E be the electrostatic energy increment due to a small change in the displacement field $\delta u(\vec{r})$ in the system. Using integration by parts, we have,

$$\begin{aligned} \delta U_E &= -\int d\vec{r}\left[\nabla_j\sigma_{ij}^M(\vec{r})\right]\delta u_i(\vec{r}) \\ &= \int d\vec{r}\sigma_{ij}^M(\vec{r})\nabla_j\delta u_i(\vec{r}). \end{aligned} \tag{29}$$

In the present case, the sample is initially tilted with the strain γ and we ask about the energy increase due to the additional strain $\delta\gamma$, i.e., $\delta\vec{u}(\vec{r}) = {}^t(\delta\gamma y,0)$. Equation (29) becomes

$$\delta U_E = \int d\vec{r}\sigma_{xy}^M(\vec{r})\delta\gamma = \delta\gamma\sigma_{xy}^M lS, \tag{30}$$

where $\sigma_{xy}^M = \Omega^{-1}\int d\vec{r}\sigma_{xy}(\vec{r})$ is the spatial average of the Maxwell stress. Using Eq. (27), we thus find

$$\sigma_{xy}^M = \frac{1}{lS}\frac{d}{d\gamma}U_E = gE_{\rm ex}^2\gamma. \tag{31}$$

The expansion of g up to a second order in $\delta\varepsilon/\bar{\varepsilon}$,

$$g = 4\bar{\varepsilon}\left[(\phi_1-\phi_1^2)\left(\frac{\delta\varepsilon}{\bar{\varepsilon}}\right)^2\right]+O\left(\left(\frac{\delta\varepsilon}{\bar{\varepsilon}}\right)^3\right) \quad \left(\frac{\delta\varepsilon}{\bar{\epsilon}}\ll 1\right), \tag{32}$$

leads to the result already obtained in Sec. III, i.e., the symmetric profile of Maxwell stress as a function of ϕ_1. As shown in Fig. 2, the profile almost agrees with that obtained through the perturbation calculation in Sec. III. This implies the relation $g = B_2K_\varepsilon$, which fixes the functional form of the factor B_2 introduced in Sec. III as

$$B_2 = 4\phi_1(1-\phi_1). \tag{33}$$

When the difference in the dielectric constants between the components becomes large, the symmetry in the Maxwell stress profile as a function of ϕ_1 is lost, as is exemplified in Fig. 2. We, however, note the relation

$$g \to \frac{\delta\varepsilon_1^2}{\bar{\varepsilon}} = K_\varepsilon \quad (\phi_1 \to 0.5). \tag{34}$$

It is possible to take into account the effect of electric conductivity in Maxwell stress. We extend our result to a weakly conductive case in the Appendix. As the result, we obtain the dependence of the shear modulus G on the frequency of the ac electric field ω as shown in Fig. 3. This dependency can be compared with the experimental result reported in Ref. [19]. Using material parameters in the experiment of Ref. [19], the characteristic frequency is obtained as $\omega_c \sim 60\ {\rm s}^{-1}$ (see the Appendix). This is comparable to the frequency where the shear modulus exhibits a sharp change in Ref. [19].

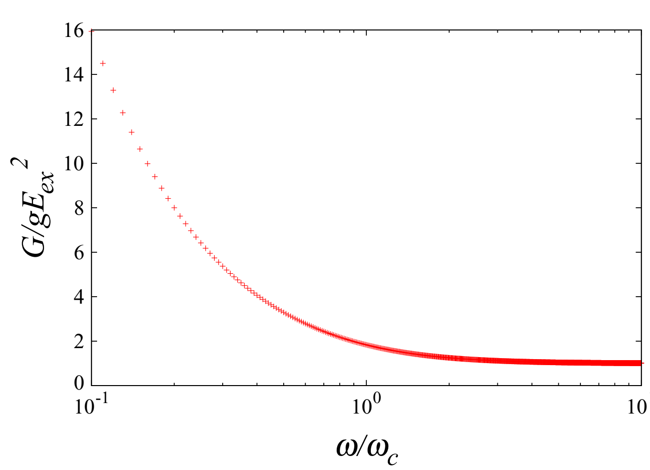

FIG. 3. Rescaled shear modulus of weakly conductive blends under ac electric field (frequency ω) calculated from Eq. (A4).

V. MAXWELL STRESS AS ANISOTROPIC INTERFACIAL TENSION

Next, we express the Maxwell stress in terms of the interfacial tensor $q_{\alpha\beta}$. Assuming the interface is flat, the angle between the interface and the electric field (y axis) ψ is defined by $\tan\psi = \gamma$. Thus, $\psi \sim \gamma$ if the shear strain γ is small enough (see Fig. 1). Since the interface is parallel to $(\sin\gamma, \cos\gamma)$, the normal vector is given by $\vec{n} = (-\cos\gamma, \sin\gamma)$ and the length of the interface is $l\sqrt{1+\gamma^2}$. To calculate the interfacial tensor, we consider a parallelogram shaped region as a unit cell. The base of the parallelogram is parallel to the x direction and its length is λ, the height is l, and the sides adjacent to the base are parallel to $\vec{e}_\parallel$ in Fig. 1. As the result, the interfacial tensor in the region is given as

$$\begin{aligned} q_{\alpha\beta} &= \frac{1}{l\lambda}\int n_\alpha n_\beta dS \\ &= \frac{2l\sqrt{1+\gamma^2}}{l\lambda} n_\alpha n_\beta \\ &= \frac{2\sqrt{1+\gamma^2}}{\lambda}\begin{pmatrix} \cos^2\gamma & -\cos\gamma\sin\gamma \\ -\cos\gamma\sin\gamma & \sin^2\gamma \end{pmatrix} \\ &\simeq \frac{2}{\lambda}\begin{pmatrix} 1 & -\gamma \\ -\gamma & 0 \end{pmatrix}. \end{aligned} \tag{35}$$

Thus, the Maxwell stress is expressed as

$$\sigma^M_{xy} = gE^2_{\rm ex}\gamma \simeq -\Gamma\frac{g\lambda}{2\Gamma}E^2_{\rm ex}q_{xy} = -\Gamma q_{xy}s_{yy}, \tag{36}$$

with a coupling tensor [17]

$$s_{\alpha\beta} = \frac{gE_{{\rm ex},\alpha}E_{{\rm ex},\beta}\lambda}{2\Gamma} = \frac{B_2K_\varepsilon E_{{\rm ex},\alpha}E_{{\rm ex},\beta}\lambda}{2\Gamma}, \tag{37}$$

where the last equality with B_2 given in Eq. (33) is valid in the small $\delta\varepsilon/\bar{\varepsilon}$ case. Since the interfacial stress is represented as Eq. (3), the total shear stress is given as

$$\sigma_{xy} = \sigma^I_{xy} + \sigma^M_{xy} = -\Gamma q_{xy}(1+s_{yy}). \tag{38}$$

Defining the shear stress as $\sigma_{xy} = G\gamma$, we obtain the shear modulus G of the system as

$$G = \frac{2\Gamma}{\lambda}(1+s_{yy}) = \frac{2}{\lambda}\Gamma + gE^2_{\rm ex}. \tag{39}$$

Equations (38) and (39) suggest that the effect of Maxwell stress on shear stress could be viewed as the renormalization of the interfacial tension with the effective value given by

$$\Gamma_{\rm eff} \equiv \Gamma(1+s_{yy}). \tag{40}$$

In the current geometry (Fig. 1), s_{yy} corresponds to the dimensionless number $\mathcal{S}$ introduced in Eq. (1) aside from the numerical constant of order unity. Therefore, in the strong electric field condition $\mathcal{S} \gg 1$, the Maxwell stress term dominates the shear modulus.

To evaluate the number $\mathcal{S}$, we need to know how the wavelength λ is selected. To this end, we recall a more general tensor form of the Maxwell stress

$$\sigma_{\alpha\beta} = -\Gamma(q_{\alpha\gamma}s_{\gamma\beta} + q_{\beta\gamma}s_{\gamma\alpha}), \tag{41}$$

which indicates that the renormalized interfacial tension is anisotropic [17]. Let us consider the phase separation process from the initial homogeneous mixture. Without the electric field, the domains grow isotropically, driven by interfacial tension. If, however, this growth process occurs under an external electric field $E_{\rm ex}$, Eqs. (38) and (41) imply an anisotropic growth due to the effective interfacial tension of the Maxwell stress origin. Supposing that the field is applied to the y direction, the stress σ_{yy} is determined by the effective interfacial tension $\Gamma(1+s_{yy})$, while only the bare interfacial tension Γ is relevant to stress σ_{xx}. This indicates the following relation for the "isotropic" growth under the electric field $\Gamma(1+s_{yy})l(x) \simeq \Gamma l(y)$, where $l(x)$ and $l(y)$ are the characteristic sizes of the domain in the x and y directions, respectively. By setting $l(x) = \lambda$ and $l(y) = l$ (the distance between plates), and assuming $s_{yy} \sim \mathcal{S} \gg 1$, we find

$$\lambda \simeq \left(\frac{\Gamma l}{gE^2_{\rm ex}}\right)^{1/2}. \tag{42}$$

This leads to

$$\mathcal{S} \simeq \left(\frac{gE^2_{\rm ex}l}{\Gamma}\right)^{1/2} \simeq \frac{l}{\lambda}. \tag{43}$$

With a different preparation condition, one may initially have completely phase-separated mixtures with a flat interface, and then apply the electric field to the direction normal to the interface. The strong enough electric field destabilizes the interface and the analysis of the fast growing mode leads to essentially the same result as Eq. (42) [22,23].

In Ref. [19], it was suggested that both components in the shear modulus (contributions from interfacial tension and Maxwell stress) are proportional to the area density of the column (in a 2D stripe structure, the analogous quantity is the line density $1/\lambda$ of the stripe). As is evident from Eq. (39), our result indicates that this does not apply to the term from the Maxwell stress. This traces back to the fact that the electrostatic interaction is long ranged. Therefore, unlike the interfacial energy, the analysis of a single column (stripe) problem multiplied by the area (line) density is not sufficient to produce the collective effect of the ensemble of columns (stripes). This remark is related to the fact that the effective interface tension defined by Eq. (40) depends on the length scale λ of the domain. Indeed, under $\mathcal{S} \gg 1$, $\Gamma_{\rm eff} \sim \lambda$, which is essential to derive the scaling law Eq. (42).

VI. SUMMARY

We have investigated the elastic property of immiscible blends under a strong electric field. Assuming a 2D periodic structure, we have calculated the Maxwell stress and interfacial stress, and derived the shear modulus expressed as Eq. (39). Under the strong field condition $\mathcal{S} \gg 1$, the Maxwell stress (second term) dominates, where the factor g represents the dependence on the composition and the dielectric properties of the blend. We have argued that essentially the same scaling form is expected for a 3D system as well, although the factor g should be refined to take into account the in-plane ordering of the columnar structures.

In addition, we have suggested the notion of an effective interfacial tension $\Gamma_{\rm eff}$ [Eq. (40)], which renormalizes the effect of the electric field. With this notion, we have derived a scaling law for the preferred column (stripe) size formed under the electric field.

To the best of our knowledge, so far, theoretical studies including our previous [18] and present works are restricted to the limit of either a weak or strong field. In the future, it should be of great interest to investigate the intermediate regime, i.e., $\mathcal{S} \sim 1$ under shear flow, where the interplay between the electric field and flow field may lead to an intriguing dynamical scenario. Another issue is related to the boundary effect. Although in our present study we implicitly neglect it, the effects associated with wetting and boundary layer formation may become increasingly relevant for blends confined in a thin region.

ACKNOWLEDGMENTS

We are grateful to H. Orihara, T. Ohta, and T. Taniguchi for many instructive discussions on the subject. This work was supported by MEXT KAKENHI Grant No. 16H00804, Fluctuation and Structure, JSPS KAKENHI Grant No. 24340100, Grant-in-Aid for Scientific Research (B), and JSPS Core-to-core Program, “Non-equilibrium dynamics of soft matter and information.”

APPENDIX

One may be interested in the electrorhological behaviors of dielectric fluid mixtures which contain a finite fraction of charge carriers. In that case, the ac electric field is often employed to avoid charge accumulation. As we mentioned in Sec. IV, our model can be extended to include the conductivity effect. When binary mixtures are conductive, Eq. (24) has to be modified. Here, we introduce the electric conductivities of components 1 and 2, κ_1 and κ_2, respectively. Then, $D_\perp$ (the component of an electric flux density perpendicular to the interface) obeys the following boundary condition at the interface,

$$D_{\perp,1} - D_{\perp,2} = \left(\frac{\varepsilon_1}{\kappa_1} - \frac{\varepsilon_2}{\kappa_2}\right) J_\perp, \tag{A1}$$

where $J_\perp = \kappa_1 E_{\perp,1} = \kappa_2 E_{\perp,2}$ is the electric current density perpendicular to the interface. The boundary condition of the parallel direction is the same as the dielectric case. By using Eq. (A1), one can rewrite Eqs. (24) and (26) so as to take into account the electric conductivity. This leads to an expression for g, which enters in the electrostatic energy of the sheared sample (see Eq. (28)):

$$\tilde{g} = -\frac{\tilde{\varepsilon}_1\tilde{\kappa}_2^2\phi_1 + \tilde{\varepsilon}_2\tilde{\kappa}_1^2\phi_2}{(\tilde{\kappa}_2\phi_1 + \tilde{\kappa}_1\phi_2)^2} + \tilde{\varepsilon}_1\phi_1 + \tilde{\varepsilon}_2\phi_2. \tag{A2}$$

Since the dielectric response generally involves the time delay under an ac field, the dielectric constants $\tilde{\varepsilon}_i = \varepsilon_i - i\kappa_i/\omega$ and the conductivities $\tilde{\kappa}_i = \kappa_i + i\omega\varepsilon_i$ become complex numbers, where ω is the frequency of the ac field. After some calculations, we obtain

$$\begin{aligned}
\tilde{g} &\equiv \mathrm{Re}[\tilde{g}] + i\,\mathrm{Im}[\tilde{g}],\\
\mathrm{Re}[\tilde{g}] &= \frac{\phi_1\phi_2}{\varepsilon_1\phi_2 + \varepsilon_2\phi_1}\left\{(\varepsilon_1 - \varepsilon_2)^2 - \left(\frac{\varepsilon_1\kappa_2 - \varepsilon_2\kappa_1}{\varepsilon_1\phi_2 + \varepsilon_2\phi_1}\right)^2 \frac{1}{\omega^2 + \omega_c^2}\right\},\\
\mathrm{Im}[\tilde{g}] &= \frac{\omega_c}{\omega}\frac{\phi_1\phi_2}{\varepsilon_1\phi_2 + \varepsilon_2\phi_1}\left\{-\left(\frac{\kappa_1 - \kappa_2}{\omega_c}\right)^2 + \left(\frac{\varepsilon_1\kappa_2 - \varepsilon_2\kappa_1}{\kappa_1\phi_2 + \kappa_2\phi_1}\right)^2 \frac{\omega^2}{\omega^2 + \omega_c^2}\right\},
\end{aligned} \tag{A3}$$

where $\mathrm{Re}[\tilde{g}]$ and $\mathrm{Im}[\tilde{g}]$ are a real part and a imaginary part of $\tilde{g}$, respectively, and $\omega_c = (\kappa_1\phi_2 + \kappa_2\phi_1)/(\varepsilon_1\phi_2 + \varepsilon_2\phi_1)$ is a characteristic frequency at which the dielectric and conductive response changes qualitatively. Modifying E_{ex} as $E_{\mathrm{ex}} \to E_{\mathrm{ex}}\exp(i\omega t)$ in Eq. (39), the shear modulus G is represented by

$$\begin{aligned}
G &= \frac{2}{\lambda}\Gamma + \left|\tilde{g}E_{\mathrm{ex}}^2 \exp(2i\omega t)\right|\\
&= \frac{2}{\lambda}\Gamma + \sqrt{(\mathrm{Re}[\tilde{g}])^2 + (\mathrm{Im}[\tilde{g}])^2}E_{\mathrm{ex}}^2.
\end{aligned} \tag{A4}$$

At high frequency $\omega \gg \omega_c$, $\mathrm{Re}[\tilde{g}]$ approaches the constant $\tilde{g}(\kappa_i = 0) = g$, while $\mathrm{Im}[\tilde{g}]$ goes to zero as $\sim 1/\omega$. Therefore, the result of the purely dielectric case Eq. (28) is recovered. On the other hand, the behavior in the low frequency limit ($\omega \ll \omega_c$) depends on the combinations of dielectric constants and conductivities of mixtures; while $\mathrm{Re}[\tilde{g}]$ approaches the constant $g - \phi_1\phi_2(\varepsilon_1\phi_2 + \varepsilon_2\phi_1)^{-1}[(\varepsilon_1\kappa_2 - \varepsilon_2\kappa_2)/(\kappa_1\phi_2 + \kappa_2\phi_1)]^2$, $\mathrm{Im}[\tilde{g}]$ diverges as $1/\omega$ in general cases (except for the $\kappa_1 = \kappa_2$ case). This indicates that the out-of-phase (conductive) response dominates in the low frequency regime, which results in the enhancement of the shear modulus. We numerically evaluate Eq. (A4) and plot it in Fig. 3, where the material parameters are adopted from the experiment in Ref. [19].

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