On diamagnetic domains in layered conductors

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(Submitted June 16, 2006)
Fiz. Nizk. Temp. **33**, 591–596 (May 2007)

The magnetic susceptibility and domain structure of a layered conductor are calculated under conditions of a substantial de Haas–van Alphen effect for arbitrary orientation of the external magnetic field with respect to the layers. It is shown that the amplitude of the magnetic susceptibility is an oscillatory function of the angle between the magnetic field and the normal to the layers, with a sharp maximum along preferred directions of magnetic field. The possibility of propagation of low-frequency weakly damped modes of the electromagnetic field is discussed with allowance for the induced magnetism of the conduction electrons. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2737556]

The possibility of an oscillatory dependence of the magnetization of metals on the value of the magnetic field at low temperatures was predicted by Landau[1] and observed by de Haas and van Alphen in 1930.[2] This oscillatory effect, due to quantization of the energy of orbital motion of electrons in the magnetic field, opened the door to a series of quantum oscillation effects, a rather complete and detailed description of which is given in the well-known monograph by Schoenberg.[3]

In the quasiclassical approximation, when the Fermi energy ε_F is much larger than the distance $\hbar\Omega$ between Landau levels (Ω is the cyclotron frequency and $\hbar$ is Planck's constant), the part magnetic susceptibility that oscillates with inverse magnetic field, $\chi_{\rm osc}$, exceeds the monotonically varying part $\chi_{\rm mon}$ by a factor of $(\varepsilon_F/\hbar\Omega)^{3/2}$ (Ref. 4). As a result, at certain sufficiently large values of the magnetic field the magnetic susceptibility χ can be greater than $1/4\pi$, and the homogeneous state of the sample becomes unstable, leading to the formation of diamagnetic domains, which were first observed by Condon.[5] The structure of the diamagnetic domains in metals was described by Condon and Walstendt[6] and by Privorotskiĭ and Azbel'[7,8] and has been investigated experimentally in some metals by resonance methods.[9–12]

Quantum oscillation effects are manifested particularly clearly in layered conductors immersed in strong external magnetic fields $\mathbf{H}_0$, since a much greater number of conduction electrons is involved in their formation than in the case of ordinary metals.[13,14] This is due to the quasi-two-dimensional character of the energy spectrum of the charge carriers in layered conductors. Their energy

$$\epsilon(\mathbf{p}) = \sum_n \epsilon_n(p_x,p_y)\cos\left(\frac{np_z}{p_0}\right) \tag{1}$$

depends weakly on the projection of the momentum on the normal to the layers, p_z, and the classical trajectories in momentum space in a magnetic field that deviates substantially from the layers are almost indistinguishable. In Eq. (1) $p_0 = \hbar/a$, where a is the distance between layers; the functions $\varepsilon_n(p_x,p_y)$ fall off with increasing index n, so that the maximum value of the function $\varepsilon_1(p_x,p_y)$ on the Fermi surface $\varepsilon(\mathbf{p})=\varepsilon_F$ can be much less than the Fermi energy, i.e.,

$$\max\epsilon_1(p_x,p_y) = \eta\epsilon_F \ll \epsilon_F. \tag{2}$$

The domain structure of layered conductors in a magnetic field orthogonal to the layers was considered by Maniv and Vagner.[15]

Let us examine the formation of diamagnetic domains in organic layered conductors for arbitrary orientation of the magnetic field with respect to the layers. The observation of Shubnikov–de Haas oscillations of the magnetoresistance of layered conductors for the most diverse orientations of the magnetic field (see, e.g., the review article by Kartsovnik[14] and the literature cited therein) indicates that at least one sheet of the Fermi surface is a slightly corrugated cylinder.

The cross section $S(\varepsilon,p_\zeta)$ of this cylinder on the $p_\zeta = (\mathbf{p}\cdot\mathbf{B})/B=$const plane depends weakly on the projection of the momentum onto the magnetic field direction:

$$\frac{\partial S(\epsilon,p_\zeta)}{\partial p_\zeta} = \sum_n I_n(\epsilon,\theta)\sin\left(\frac{np_\zeta}{p_0\cos\theta}\right) \sim \eta S(\epsilon,p_\zeta)/p_0. \tag{3}$$

For some orientations of the magnetic field, any of the coefficients $I_n(\varepsilon,\theta)$ of the Fourier series in the linear approximation in the quasi-two-dimensionality parameter of the energy spectrum, η, can vanish. For those values of the angle θ between the normal to the layers and the magnetic field vector at which the largest of the coefficients $I_1(\varepsilon,\theta)$ vanishes, the amplitude of the quantum oscillations of the magnetic susceptibility and magnetoresistance increases sharply,[16] providing the most favorable conditions for observation of the domain structure.

The magnetic field that acts on the charge carriers is the value averaged over a region of the order of the Larmor radius, i.e., the magnetic induction $\mathbf{B}$. As long as the magnetic susceptibility is small, one can neglect the difference between $\mathbf{B}$ and $\mathbf{H}$. However, at sufficiently low temperatures the magnetic susceptibility can reach values of the order of unity, and the magnetization $\mathbf{M}(\mathbf{B})$ and the magnetic field $\mathbf{H}=\mathbf{B}-4\pi\mathbf{M}(\mathbf{B})$ become complicated functions of the magnetic induction, and for $\chi > 1/4\pi$ the sample breaks up into alternating domains with different values of the induction.[5,7]

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The magnetic induction in the conductor can be represented in the form $\mathbf{B}(y)=\mathbf{B}_0+\mathbf{B}_1(y)$, where $\mathbf{B}_1(y)$ is the nonuniform contribution, which satisfies the equation

$$\text{curl}\,\mathbf{B}_1=\frac{4\pi}{c}\mathbf{j}^{(m)}. \tag{4}$$

Let us find the relation between the current density $\mathbf{j}^{(m)}=c\,\text{curl}\mathbf{M}$ induced by an external magnetic field with induction $\mathbf{B}_0=(B_0\sin\theta,0,B_0\cos\theta)$, where θ is the angle between the magnetic field and the normal to the layers (the z axis), and c is the speed of light. The linearized kinetic equation for the density matrix has the form[17,18]

$$\frac{i}{\hbar}[\hat{H}_0,\hat{\rho}_1]+\frac{i}{\hbar}[\hat{H}_1,\hat{\rho}_0]=\hat{I}(\hat{\rho}_1). \tag{5}$$

Here $\hat{\rho}_0$ and $\hat{H}_0$ are the equilibrium density matrices and Hamiltonian in the one-electron approximation, $\hat{\rho}_1$ and $\hat{H}_1$ are the nonuniform additions to them, and $\hat{I}(\hat{\rho}_1)\sim\tau^{-1}\hat{\rho}_1$ is the collision operator, the eigenvalues of which coincide in order of magnitude with the inverse mean free time of the charge carriers. Under conditions when it is important to take the quantization of the electron energy levels into account, the cyclotron frequency Ω is much greater than the collision frequency $(\Omega\gg\tau^{-1})$, and the magnetization current density can be written in the form

$$\mathbf{j}^{(m)}=\text{Tr}(\hat{\mathbf{j}}\hat{\rho})=\sum_{\alpha,\alpha'}\frac{w_\alpha-w_{\alpha'}}{\epsilon_\alpha-\epsilon_{\alpha'}}H_{1\alpha,\alpha'}(\mathbf{r})\mathbf{j}_{\alpha',\alpha}(\mathbf{r})-\frac{e^2}{mc}\mathbf{A}_1(\mathbf{r})\sum_\alpha w_\alpha|\Psi_\alpha|^2, \tag{6}$$

where $\alpha=n,p_y,p_\zeta,\sigma$ is a set of quantum quantum numbers characterizing the state of the conduction electron. The ζ axis in the coordinate system ξ,y,ζ is directed along the vector $\mathbf{B}_0$, $p_\zeta=p_z\cos\theta-p_x\sin\theta=(\mathbf{p}\cdot\mathbf{B}_0)/p$, $\sigma=\pm1$ is the projection of the electron spin, $w_\alpha=[\exp(\varepsilon_\alpha-\mu)+1]^{-1}$ is the occupation probability of the state with quantum numbers n,p_y,p_ζ,σ, $\varepsilon_\alpha=\varepsilon_{n,p_y,p_\zeta}-\mu_B B_0\sigma$,

$$\hat{H}_1(\mathbf{r})=-\frac{1}{c}\int d^3r'\hat{\mathbf{A}}_1\hat{\mathbf{j}}(\mathbf{r},\mathbf{r}') \tag{7}$$

is the correction to the Hamiltonian, μ is the chemical potential, μ_B is the Bohr magneton,

$$\hat{\mathbf{j}}(\mathbf{r},\mathbf{r}')=\frac{e}{2}\left[\hat{\mathbf{v}}\left(\hat{\mathbf{p}}-\frac{e}{c}\mathbf{A}_0(y)\right)\delta(\mathbf{r}-\mathbf{r}')+\delta(\mathbf{r}-\mathbf{r}')\hat{\mathbf{v}}\left(\hat{\mathbf{p}}-\frac{e}{c}\mathbf{A}_0(y)\right)\right] \tag{8}$$

is the current density operator, and $\mathrm{v}(\mathbf{p})=\partial\varepsilon(\mathbf{p})/\partial p$.

The vector potential is conveniently chosen in the Landau gauge:

$$\mathbf{A}(\mathbf{r})=(A(y),0,0),$$

$$A(y)=B_0y+\int_0^y dyB_1(y)\equiv A_0(y)+A_1(y). \tag{9}$$

The matrix elements $H_{1\alpha,\alpha'}$ and $\mathbf{j}_{\alpha,\alpha'}(\mathbf{r})$ are calculated in the basis of eigenfunctions ψ_α of the unperturbed Hamiltonian, which satisfy the equation

$$\hat{\epsilon}\left(\hat{\mathbf{p}}-\frac{e}{c}\mathbf{A}_0(y)\right)\Psi_\alpha=\epsilon_\alpha\Psi_\alpha. \tag{10}$$

For determination of the character of the domain structure in layered conductors it is not so important to take into account the anisotropy of the energy spectrum of the charge carriers in the plane of the layers, and for the sake of brevity in the calculations we shall keep only the first two terms in the Eq. (1) for the energy of the charge carriers, and we shall also assume that $\varepsilon_0(p_x,p_y)$ is an isotropic function and that $\varepsilon_1(p_x,p_y)$ is a constant, denoted by ε_0, i.e., we use a rather simple dispersion relation for the conduction electrons:

$$\epsilon(\mathbf{p})=\frac{p_x^2+p_y^2}{2m}-\epsilon_0\cos\frac{p_z}{p_0}, \tag{11}$$

where m is the effective mass of the electrons in the plane of the layers.

In the leading approximation in the small parameter $\eta=\varepsilon_0/\varepsilon_F$ the electron is a harmonic oscillator with frequency $\Omega_0=|e|B_0\cos\Psi/mc$. Expanding the nonuniform correction to the vector potential into a Fourier integral,

$$A_1(y)=\int dkA_1(k)e^{iky},$$

we obtain the following expression for the magnetization current density:

$$\mathbf{j}^{(m)}=-c\frac{\chi_0}{a_B^2}\sum_{n,n',\sigma}\int dp_\zeta\frac{w_{np\zeta\sigma}-w_{n'p\zeta\sigma}}{n-n'}\int dkA_1(k)e^{iky}\left|\langle n|e^{iqu}|n'\rangle\right|^2-c\frac{\chi_0}{a_B^2}\mathbf{A}_1(\mathbf{r})\sum_{n,\sigma}\int dp_\zeta w_{np\zeta\sigma}, \tag{12}$$

where

$$\chi_0=\frac{1}{(2\pi)^2}\frac{e^2}{mc^2}\frac{p_0}{\hbar},\quad a_B=\sqrt{\frac{\hbar}{m\Omega_0}},$$

$$|n\rangle=\frac{1}{\pi^{1/4}\sqrt{2^n n!}}\exp\left(-\frac{u^2}{2}\right)H_n(u),$$

$H_n(u)$ is an Hermite polynomial, $u=(y-y_0)/a_B$, and $y_0=e(p_\xi+p_\zeta\tan\theta)/(cB_0)$. The matrix element

$$\langle n|e^{iqu}|n+p\rangle=\sqrt{\frac{n!}{(n+p)!}}i^p\left(\frac{q^2}{2}\right)^{\frac{p}{2}}e^{\frac{q^2}{4}}L_n^p(p^2/2),$$

where $q=a_Bk$ is expressed in terms of the associated Laguerre polynomials $L_n^p(q^2/2)$.

In the case of a weak inhomogeneity $kr_0\ll 1$ the matrix element can be expanded in powers of q, and the linear magnetization current density takes the form

$$j_\xi^{(m)}=c\chi\frac{\partial B_\zeta}{\partial y}+c\Lambda r_0^2\frac{\partial^3 B_\zeta}{\partial y^3},\tag{13}$$

where

$$\chi=-\chi_0\sum_{n\sigma}\int_{-\pi}^{\pi}d\beta\frac{\partial}{\partial n}(n_1^2 w_{np\zeta\sigma}),$$

$$\Lambda=-\frac{\chi_0}{4}\sum_{n\sigma}\int_{-\pi}^{\pi}d\beta\frac{\partial}{\partial n}\left[n_1\left(n_1^2+\frac{1}{4}\right)w_{np\zeta\sigma}\right],$$

$$\beta=\frac{p_\zeta}{p_0\cos\theta},\quad r_0=\frac{v_F}{\Omega_0},\quad n_1=n+\frac{1}{2}.\tag{14}$$

The peculiarities of the quasi-two-dimensional electron energy spectrum of layered conductors lead to oscillatory dependence of the amplitude of the magnetic susceptibility on the angle between the magnetic field and the normal to the layers. In the linear approximation in the quasi-two-dimensionality parameter η the areas of the electron orbits in momentum space corresponding to the spectrum (11),

$$S(\epsilon,p_\zeta)=\frac{2\pi m\epsilon}{\cos\theta}\left(1+\frac{\epsilon_0}{\epsilon}J_0(\alpha)\cos\beta\right),\tag{15}$$

where $\alpha=(\sqrt{2m\varepsilon}/p_0)\tan\theta$, are identical when the Bessel function $J_0(\alpha)$ vanishes.[19] For such orientations of the vector $\mathbf{B}_0$ the amplitudes of the oscillatory (in inverse magnetic field) parts of the kinetic and thermodynamic characteristics of the conductor increase substantially.

Using the Poisson summation formula and the quasiclassical quantization rules

$$S(\epsilon,p_\zeta)=\frac{2\pi|e|B_0\hbar}{c}\left(n+\frac{1}{2}\right),$$

we find that, under the condition $\eta^2\varepsilon_F\ll\hbar\Omega\ll\eta\varepsilon_F$, the expressions for χ and Λ become

$$\chi=-\frac{2}{\pi}\frac{e^2}{mc^2}\frac{p_0}{\hbar}\left(\frac{\mu}{\hbar\Omega_0}\right)^2\times\sum_{l=1}^{\infty}(-1)^l J_0(l\Delta(\mu))\Psi(\lambda l)\cos\frac{2\pi l\mu}{\hbar\Omega_0}\cos\frac{\pi lm}{m_e\cos\theta},\tag{16}$$

$$\Lambda=\frac{1}{4}\left(\frac{\mu}{\epsilon_F}\right)\chi.\tag{17}$$

Here

$$\Psi(z)=\frac{z}{\sinh z},\quad \lambda=\frac{2\pi^2T}{\hbar\Omega_0},\quad \Delta(\mu)=\frac{2\pi\epsilon_0}{\hbar\Omega_0}J_0(\alpha(\mu)),$$

m_e is the mass of the free electron. Neglecting the quantum oscillations of the chemical potential, we set $\mu=\varepsilon_F$.

The amplitude of the oscillatory part of the magnetic susceptibility $\chi_m\sim\chi_0(\varepsilon_F/\hbar\Omega_0)^2J_0(\Delta(\mu))$ for $\Delta(\mu)\gg 1$ is equal in order of magnitude to $\chi_0(\varepsilon_F/\hbar\Omega_0)^2\sqrt{\hbar\Omega_0/\eta\varepsilon_F}$. For the values $\theta=\theta_i$ at which $\alpha_i=(\sqrt{2m\varepsilon_F}/p_0)\tan\theta_i$ is a root of the Bessel function $J_0(\alpha_i)=0$, the amplitude χ_m grows to values of the order of $\chi_0(\varepsilon_F/\hbar\Omega_0)^2$. At these orientations of the magnetic field the dependence of the areas of section of the Fermi surface $S(\varepsilon,p_\zeta)$ on the momentum projection p_ζ are manifested only in the terms quadratic in η, i.e., $\partial S(\varepsilon,p_\zeta)/\partial p_\zeta\sim O(\eta^2)$.

When $\kappa^2\equiv|1-4\pi\chi(B_0)|\ll 1$, the linear term of the expansion of $\mathbf{j}^{(m)}$ in powers of $\mathbf{B}_1(y)$ can be of the same order of magnitude as the nonlinear term proportional to $B_1^3(y)$. As a result, the magnetization current density may be written in the form

$$j_\xi^{(m)}=c\frac{\partial M_\zeta}{\partial y}=c\chi\frac{\partial B_1}{\partial y}+c\Lambda r_0^2\frac{\partial^3 B_1}{\partial y^3}-c\Gamma\frac{\partial B_1^3}{\partial y},\tag{18}$$

where $\Gamma=-\partial^2M(B_0)/\partial B_0^2\simeq(\varepsilon_F/\hbar\Omega_0B_0)^2$.

Maxwell's Eq. (4) has the solution

$$B_1(y)=b_0\frac{s}{\sqrt{1+s^2}}\mathrm{sn}\left(\frac{y}{\delta\sqrt{1+s^2}},s\right),\tag{19}$$

describing a periodic domain structure with period $Y=4\delta\sqrt{1+s^2}K(s)$ and a domain-wall thickness $\delta=\sqrt{\pi r_0/2\kappa}$. Here $b_0=(\kappa^2/2\pi\Gamma)^{1/2}\approx\kappa B_0(\hbar\Omega/\varepsilon_F)$, and $K(s)$ is the complete elliptic integral of the first kind. The modulus s of the Jacobi elliptic function sn determines the period Y and is found from the condition of minimum (with respect to Y) total thermodynamic potential, including the surface energy at the domain walls. In the most important case in practice, when the linear dimensions L of the sample are much greater than the Larmor radius of the electron, $Y\sim\sqrt{\kappa^2r_0L}$ (Ref. 8). Without loss of generality, it can be assumed that the size of the domains is large compared to δ.

Experiments on the propagation of electromagnetic waves in a sample of finite dimensions allow one to trace the formation of the domain structure and to determine its parameters. In the case of weak temporal and spatial dispersion, i.e.,

$$\omega\tau\ll 1,\quad kr_0\ll 1,\quad \eta k_z v_F\tau\ll 1,\tag{20}$$

where ω and k are the frequency and wave vector of the ac field, the system can be tuned to the instantaneous values of the ac fields, and one can use the dc expressions for the current density and magnetization, plugging in the values of the ac fields at the given instant. In the expressions for the conduction current density one can neglect the gradient terms proportional to powers of the small parameter $(kr_0)^2$. As a result, one can obtain the following equation for the ac fields $\mathbf{B}^{\sim}$ and $\mathbf{H}^{\sim}$:

$$\frac{\partial \mathbf{B}}{\partial t} = -\frac{c^2}{4\pi}\mathrm{curl}(\hat{\rho}\,\mathrm{curl}\,\mathbf{H}), \quad (21)$$

where $(\hat{\rho}\,\mathrm{curl}\,\mathbf{H})_i = \rho_{ik}(\mathrm{curl}\,\mathbf{H})_k$. For $\theta \neq \theta_i$ the quantum corrections to the conductivity tensor, proportional to the small parameter $(\hbar\Omega_0/\eta\varepsilon_F)^{1/2}$, are usually small compared with the smoothly varying part, and the resistivity tensor corresponding to the dispersion relation (11) in a coordinate system ξ, y, ζ whose ζ axis is directed along the external magnetic field can be written in the form

$$\rho_{ik} = \begin{pmatrix} \dfrac{\sigma_{zz}\cos^2\theta + \sigma_0\sin^2\theta}{\sigma_0\sigma_{zz}} & -\dfrac{\Omega\tau}{\sigma_0\cos\theta} & \dfrac{\cos\theta\sin\theta(\sigma_{zz}-\sigma_0)}{\sigma_0\sigma_{zz}} \\ \dfrac{\Omega\tau}{\sigma_0\cos\theta} & \sigma_0^{-1} & 0 \\ \dfrac{\cos\theta\sin\theta(\sigma_{zz}-\sigma_0)}{\sigma_0\sigma_{zz}} & 0 & \dfrac{\sigma_{zz}\sin^2\theta + \sigma_0\cos^2\theta}{\sigma_0\sigma_{zz}} \end{pmatrix}, \quad (22)$$

where σ_0 and $\sigma_{zz} \sim \eta^2\sigma_0$ are the conductivities in the plane of the layers and along the normal to the layers, respectively, in the absence of external magnetic field.

When the wave vector $k=(0,k_y,k_\zeta)$ lies in the $y\zeta$ plane, the equation for the field component $B_\zeta^{\sim}(y,\zeta,t)$ has the form

$$\left[\frac{\partial}{\partial t} - \frac{c^2}{4\pi}\left(\rho_{22}\frac{\partial^2}{\partial\zeta^2} + \rho_{33}\frac{\partial^2}{\partial y^2}\right)\right]\frac{\partial B_\zeta^{\sim}}{\partial t} = \left[\frac{c^2}{4\pi}\rho_{11}\frac{\partial}{\partial t} - \left(\frac{c^2}{4\pi}\right)^2\left((\rho_{11}\rho_{33} - \rho_{13}^2)\frac{\partial^2}{\partial y^2} + (\rho_{11}\rho_{22} + \rho_{12}^2)\frac{\partial^2}{\partial\zeta^2}\right)\right]\left(\frac{\partial^2 H_\zeta^{\sim}}{\partial y^2} + \frac{\partial^2 B_\zeta^{\sim}}{\partial\zeta^2}\right), \quad (23)$$

where

$$H_\zeta^{\sim} = (1-4\pi\chi)B_\zeta^{\sim} - 4\pi\Lambda r_0^2\frac{\partial^2 B_\zeta^{\sim}}{\partial y^2} + 4\pi\Gamma B_\zeta^{\sim -3}. \quad (24)$$

For $1-4\pi\chi>0$ in the linear approximation in the small wave amplitude the solution of Eq. (23) can be sought in harmonic form, assuming $B_\zeta^{\sim}, H_\zeta^{\sim} \sim \exp(-i\omega t + i\mathbf{k}\cdot\mathbf{r})$. As a result, we obtain the following dispersion relation:

$$\omega^2 - \left(\frac{c^2}{4\pi}\right)^2[(\rho_{11}\rho_{33} - \rho_{13}^2)k_y^2 + (\rho_{11}\rho_{22} + \rho_{12}^2)k_\zeta^2]$$
$$\times[k_\zeta^2 + (1-4\pi\chi)k_y^2 + 4\pi\Lambda r_0^2 k_y^4] + i\omega\frac{c^2}{4\pi}[k_\zeta^2(\rho_{11}+\rho_{22})$$
$$+ ((1-4\pi\chi)\rho_{11} + \rho_{33})k_y^2 + 4\pi\Lambda\rho_{11}r_0^2 k_y^4] = 0. \quad (25)$$

Under the condition $\eta^{-2} \gg \Omega\tau$, which holds for quasi-two-dimensional conductors, the imaginary part of the dispersion relation (25), generally speaking, is of the same order as the real part, and the eigenmodes are strongly damped. An exception is the case of small θ and k_y, specifically: $\theta < \eta^2$, $k_y < \eta^2 k_\zeta$. Then it is possible for a helicoidal wave to propagate, with the frequency

$$\omega = \frac{c^2}{4\pi}\rho_{12}k^2 - i\left(\frac{c^2}{8\pi}\right)(\rho_{11}+\rho_{22})k^2. \quad (26)$$

Under conditions for which domain structure exists, i.e., for $1-4\pi\chi<0$, the time-varying field along the OY axis is nonuniform, leading to strong damping of the wave.

In layered conductors with an arbitrary electron energy spectrum (1) the maxima in the angular dependence of the amplitudes of the de Haas–van Alphen and Shubnikov–de Haas oscillations are not as sharp, since the coefficients in formula (3) do not vanish simultaneously for any orientation of the vector $\mathbf{B}_0$ (Ref. 20), although the conditions for observation of domain structure are undoubtedly more favorable than for metals in which the charge carriers have a quasi-isotropic energy.

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Translated by Steve Torstveit