

# Solving the Ultranonlocality Problem in Time-Dependent Spin-Density-Functional Theory

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(Received 10 September 2002; published 11 February 2003)

It has been known for some time that the exchange-correlation potential in time-dependent density-functional theory is an intrinsically nonlocal functional of the density as soon as one goes beyond the adiabatic approximation. In this paper we show that a much more severe nonlocality problem, with a completely different physical origin, plagues the exchange-correlation potentials in time-dependent spin-density functional theory. We show how the use of the *spin current density* as a basic variable solves this problem, and we provide an explicit local expression for the exchange-correlation fields as functionals of the spin currents.

DOI: 10.1103/PhysRevLett.90.066402

PACS numbers: 71.15.Mb, 71.10.Ca, 71.45.Gm

For many years the local density approximation (LDA) has provided the much needed handle on the difficult problem of approximating the density dependence of the exchange-correlation (xc) potential—the single particle potential that incorporates the many-body effects in the Kohn-Sham equation for the ground state density [1]. In LDA, the xc potential  $V_{xc}(\vec{r})$  is simply a function of the local density  $n(\vec{r})$ . This approximation is not unreasonable as long as the functional derivative of  $V_{xc}(\vec{r})$  with respect to  $n(\vec{r}')$ —the so-called *exchange-correlation kernel*  $f_{xc}(\vec{r}, \vec{r}') \equiv [\delta V_{xc}(\vec{r})/\delta n(\vec{r}')]—$ is a sufficiently short-ranged function of the distance  $|\vec{r} - \vec{r}'|$  [2].

However, much recent work [3–7] has demonstrated that the requirement of short rangedness is not always fulfilled in physical systems, and when this happens the local density approximation is flawed. This does not mean that a local description of exchange and correlation is absolutely impossible, only that such a description cannot be achieved in terms of the particle density.

For example, in the density-functional theory of crystalline insulators it has been found [4–6] that the xc potential has an “ultranonlocal” dependence on the density, due to the fact that the Fourier transform of the xc kernel  $f_{xc}(\vec{k}, \vec{k})$  diverges as  $1/k^2$  for  $k \rightarrow 0$ . But, the ultranonlocality disappears if one reformulates the theory in terms of the electric polarization  $\vec{P}(\vec{r})$  and the exchange-correlation electric field  $\vec{E}_{xc}(\vec{r})$  associated with it.

Another instance of the ultranonlocality problem was discovered in the time-dependent density-functional theory (TDDFT) [8] following the realization that the frequency-dependent LDA [9] fails to satisfy Kohn’s theorem [10,11]. The pathology was traced to a singularity of the form  $\frac{\vec{k} \cdot \vec{k}'}{k^2}$  in the xc kernel  $f_{xc}(\vec{k}, \vec{k}', \omega)$  for  $k \rightarrow 0$  at finite  $\vec{k}'$  and  $\omega$ . The ensuing nonlocality problem was solved by upgrading to time-dependent *current*-density functional theory (TDCDFT), where the basic variable is the current density, and its conjugate field is a vector potential [3]. TDCDFT has since been applied to the calculation of the optical spectra of solids [12] and the

polarizability of long polymer chains [13] with considerable success.

In this Letter we show that the ultranonlocality problem occurs in an *aggravated form* in the time-dependent spin-density functional theory or, more generally, in the time-dependent DFT of any multi-component system. The novel features of the spin-dependent problem stem from the fact that the xc kernel presents a divergence even in the homogeneous electron liquid. More precisely, it can be shown that the Fourier transform of the spin-dependent exchange-correlation kernel  $f_{xc,\sigma\sigma'}(r - r', t - t') \equiv [\delta V_{xc,\sigma}(\vec{r}, t)/\delta n_{\sigma'}(\vec{r}', t')]$  in a homogeneous electron liquid has the long-wavelength expansion

$$f_{xc,\sigma\sigma'}(k, \omega) \xrightarrow{k \rightarrow 0} \frac{A(\omega)}{k^2} \frac{\sigma\sigma' n^2}{4n_{\sigma}n_{\sigma'}} + B_{\sigma\sigma'}(\omega) + O(k^2), \quad (1)$$

where  $A(\omega)$  and  $B_{\sigma\sigma'}(\omega)$  are complex functions of frequency,  $n_{\sigma}$  is the density of  $\sigma$ -spin electrons ( $\sigma = +1$  for  $\uparrow$ -spin and  $\sigma = -1$  for  $\downarrow$ -spin), and  $n = n_{\uparrow} + n_{\downarrow}$  is the total density. Since the xc potential created by a small density variation  $\delta n_{\sigma}(\vec{k}, \omega)$  is given by the formula

$$V_{xc,\sigma}(\vec{k}, \omega) = \sum_{\sigma'} f_{xc,\sigma\sigma'}(k, \omega) \delta n_{\sigma'}(\vec{k}, \omega), \quad (2)$$

we see that Eq. (1) rules out the possibility of a local connection between  $V_{xc,\sigma}(\vec{r}, t)$  and  $\delta n_{\sigma'}(\vec{r}', t')$ .

The existence of the long-wavelength singularity in  $f_{xc,\sigma\sigma'}(k, \omega)$  has been known for some time. It was first pointed out by Goodman and Sjölander [14] that the third-moment sum rule for the spin-density response function implies such a singularity. Approximate formulae for  $f_{xc,-}(k, \omega) = f_{xc,\uparrow}(k, \omega) - f_{xc,\downarrow}(k, \omega)$  exhibiting the singularity were proposed in [15] and, for imaginary frequencies, in [16]. More recently, D’Amico and Vignale [17] have shown that, at low frequency and finite temperature, the singularity is related to the friction that arises between up- and down-spin currents when they

have different average velocities (the so-called spin-drag effect).

By contrast, the implications of Eq. (1) for spin-density functional theory have not been explored so far. This is understandable, since the singularity (1) arises only at finite frequency [ $A(0) = 0$ ] and therefore does not affect the *static* spin DFT. Furthermore, the singularity does not show up in the density response of spin-compensated systems, since the relevant combination of xc kernels, in that case, is  $\sum_{\sigma\sigma'} n_{\sigma} n_{\sigma'} f_{xc,\sigma\sigma'}$ , which is nonsingular. It is only in the time-dependent spin DFT [18] that the issue of the singularity becomes really critical not only to the calculation of the spin-response, but even to the calculation of just the density response[19].

In this Letter we propose a resolution of the ultra-nonlocality problem based on the use of the spin components of the current density  $\vec{j}_{\uparrow}(\vec{r}, \omega)$  and  $\vec{j}_{\downarrow}(\vec{r}, \omega)$  as basic variables. We provide an explicit expression for the spin-dependent exchange-correlation field  $\vec{E}_{xc,\sigma}(\vec{r}, \omega)$  as a local linear functional of the currents  $\vec{j}_{\sigma}$ .

The general method for upgrading from the density to the current-density formulation is described in detail in Ref. [20], so we mention only the essential steps here. We introduce a spin-dependent xc vector potential  $\vec{A}_{xc,\sigma}(\vec{k}, \omega)$  [whose time derivative,  $i\omega\vec{A}_{xc,\sigma}(\vec{k}, \omega) = \vec{E}_{xc}(\vec{k}, \omega)$ , is the xc electric field], and notice that this is linearly related to the currents in the following manner

$$A_{xc,\sigma}^{\alpha}(\vec{k}, \omega) = \frac{k^2}{\omega^2} \sum_{\sigma'} f_{xc,\sigma\sigma'}^{\alpha}(\vec{k}, \omega) j_{\sigma'}^{\alpha}(\vec{k}, \omega), \quad (3)$$

where the superscript  $\alpha$  denotes the longitudinal ( $\alpha = L$ ) or transverse ( $\alpha = T$ ) component of a vector relative to the direction of  $\vec{k}$ . It is not difficult to see that the *longitudinal* xc kernel defined in this manner coincides with the xc kernel of Eq. (1). The extra factor  $k^2/\omega^2$  in Eq. (3) exactly cancels the small- $k$  singularity of  $f_{xc}$ , and leads to a theory that admits a local approximation. The imaginary part of the current xc kernel  $f_{xc,\sigma\sigma'}^{\alpha}(k, \omega)$  is expressed in terms of a causal response function as follows:

$$\text{Im}f_{xc,\sigma\sigma'}^{\alpha}(k, \omega) = \frac{1}{V n_{\sigma} n_{\sigma'} k^2} \text{Im} \langle \langle \hat{F}_{\sigma}^{\alpha}(\vec{k}); \hat{F}_{\sigma'}^{\alpha}(-\vec{k}) \rangle \rangle_{\omega}, \quad (4)$$

where  $\langle \langle \hat{A}; \hat{B} \rangle \rangle_{\omega} \equiv -\frac{i}{\hbar} \int_0^{\infty} [\hat{A}(t), \hat{B}] e^{i\omega t} dt$  is the linear response function associated with the operators  $\hat{A}$  and  $\hat{B}$ ;  $\hat{F}_{\sigma}^{\alpha}(\vec{k}) = -\frac{im}{\hbar} [\hat{H}, \hat{j}_{\sigma}^{\alpha}(\vec{k})]$  is the time derivative of the Fourier transform of the current-density operator  $\hat{j}_{\sigma}^{\alpha}(\vec{k})$ ,  $\hat{H}$  is the Hamiltonian, and  $V$  is the volume.

Once the imaginary part of  $f_{xc,\sigma\sigma'}^{\alpha}(k, \omega)$  is known, its real part is determined by the Kramers-Krönig dispersion relation

$$\begin{aligned} \text{Re}f_{xc,\sigma\sigma'}^{\alpha}(k, \omega) &= f_{xc,\sigma\sigma'}^{\alpha}(k, \infty) \\ &\quad - \frac{2}{\pi} \mathcal{P} \int_0^{\infty} d\omega' \frac{\omega' \text{Im}f_{xc,\sigma\sigma'}^{\alpha}(k, \omega')}{\omega^2 - \omega'^2}, \end{aligned} \quad (5)$$

where  $\mathcal{P}$  denotes the principal part integral, and the infinite frequency limit of  $f_{xc,\sigma\sigma'}$  is determined by the *third moment sum rule*. In a three-dimensional electron liquid, this sum rule gives

$$\begin{aligned} f_{xc,\sigma\sigma'}^{\alpha}(k, \infty) &\xrightarrow{k \rightarrow 0} -\frac{4\pi e^2}{3k^2} \frac{n_{\uparrow} n_{\downarrow}}{n_{\sigma} n_{\sigma'}} [g_{\uparrow\downarrow}(0) - 1] \sigma\sigma' \\ &\quad + a^{\alpha} \frac{t_{c\sigma}}{n_{\sigma}} \delta_{\sigma\sigma'} + \frac{b^{\alpha}}{2} \int d\vec{r} \frac{e^2}{r} [g_{\sigma\sigma'}(r) - 1], \end{aligned} \quad (6)$$

where  $a^L = 2$ ,  $a^T = 2/3$ ,  $b^L = 4/15$ , and  $b^T = -2/15$ . Here  $g_{\sigma\sigma'}(r)$  is the spin-resolved pair correlation function and  $t_{c\sigma}$  is the average correlation kinetic energy of the  $\sigma$ -spin component. Note that the result for the longitudinal case was first obtained in Ref. [14].

It is evident from the above equations that both the longitudinal and the transverse kernels exhibit  $1/k^2$  singularities, which are “cured” by the  $k^2/\omega^2$  factor of Eq. (3). In particular, substituting the small- $k$  expansion  $\hat{F}_{\sigma}^{\alpha}(\vec{k}) = \hat{F}_{\sigma}^{\alpha}(0) + O(\vec{k})$  in Eq. (4), where  $\hat{F}_{\sigma}^{\alpha}(0)$  is the operator of the total force acting on  $\sigma$ -spin electrons, and noting that terms of first order in  $\vec{k}$  vanish by inversion symmetry, we see that the xc kernels have the small- $k$  expansion

$$f_{xc,\sigma\sigma'}^{\alpha}(k, \omega) \xrightarrow{k \rightarrow 0} \frac{A(\omega)}{k^2} \frac{\sigma\sigma' n^2}{4n_{\sigma} n_{\sigma'}} + B_{\sigma\sigma'}^{\alpha}(\omega) + O(k^2), \quad (7)$$

where

$$\text{Im}A(\omega) = -\frac{4}{V n^2} \text{Im} \langle \langle \hat{F}_{\uparrow}^{\alpha}; \hat{F}_{\downarrow}^{\alpha} \rangle \rangle_{\omega} \quad (8)$$

and

$$\begin{aligned} \text{Re}A(\omega) &= -\frac{16\pi e^2}{3} [g_{\uparrow\downarrow}(0) - 1] \\ &\quad - \frac{2}{\pi} \mathcal{P} \int_0^{\infty} d\omega' \frac{\omega' \text{Im}A(\omega')}{\omega^2 - \omega'^2}. \end{aligned} \quad (9)$$

The factor  $\sigma\sigma'$  in Eq. (7) arises from the fact that the total force  $\hat{F}_{\uparrow} + \hat{F}_{\downarrow}$  vanishes, due to translational invariance, so that  $\langle \langle \hat{F}_{\sigma}; \hat{F}_{\sigma'} \rangle \rangle_{\omega} = -\sigma\sigma' \langle \langle \hat{F}_{\uparrow}; \hat{F}_{\downarrow} \rangle \rangle_{\omega}$ . Notice also that  $A(\omega)$  is independent of the direction  $\alpha$ —longitudinal or transverse. The microscopic expression for  $B_{\sigma\sigma'}^{\alpha}$  is more complicated: a simple approximation for this quantity will be presented below.

Substituting the expansion (7) into Eq. (3), calculations similar to those described in [20] lead us to the following *local* approximation for the xc field in terms of the spin currents

$$-e\vec{E}_{xc,\sigma}(\omega) = -\vec{\nabla}V_{xc,\sigma}^{LDA} + \frac{1}{n_\sigma}\vec{\nabla}\cdot\vec{\sigma}_{xc,\sigma}(\omega) + \frac{in^2A(\omega)}{4\omega}\sum_{\sigma'}\frac{\sigma\sigma'}{n_\sigma n_{\sigma'}}\vec{j}_{\sigma'}. \quad (10)$$

Here the  $\vec{r}$  dependence has been left implicit, and the xc stress tensor  $\vec{\sigma}_{xc}(\omega)$ , as well as  $A(\omega)$ , is a function of the local spin densities, as discussed below.

Equation (10) is the central result of this paper. The first two terms on the right are well known: they are, respectively, the adiabatic LDA contribution and the viscoelastic force term, where the stress tensor  $\sigma_{xc,\sigma}(\omega)$  is related to  $B_{xc,\sigma\sigma'}$  by obvious extensions of the formulas reported in [20]. The expression for the xc stress tensor is

$$\sigma_{xc,\sigma,ij} = \sum_{\sigma'}\left[\eta_{xc,\sigma\sigma'}\left(\frac{\partial u_{\sigma',i}}{\partial r_j} + \frac{\partial u_{\sigma',j}}{\partial r_i} - \frac{2}{3}\vec{\nabla}\cdot\vec{u}_{\sigma'}\delta_{ij}\right) + \zeta_{xc,\sigma\sigma'}\vec{\nabla}\cdot\vec{u}_{\sigma'}\delta_{ij}\right], \quad (11)$$

where  $\vec{u}_\sigma = \vec{j}_\sigma/n_\sigma$ , and

$$\eta_{xc,\sigma\sigma'} = -\frac{n_\sigma n_{\sigma'}}{i\omega}B_{\sigma\sigma'}^T(\omega), \quad (12)$$

$$\zeta_{xc,\sigma\sigma'} = -\frac{n_\sigma n_{\sigma'}}{i\omega}\left[B_{\sigma\sigma'}^L(\omega) - \frac{4}{3}B_{\sigma\sigma'}^T(\omega) - \epsilon_{xc,\sigma\sigma'}''\right], \quad (13)$$

where  $\epsilon_{xc,\sigma\sigma'}'' = (\partial^2\epsilon_{xc}/\partial n_\sigma\partial n_{\sigma'})$ . The last term in Eq. (10) is new, and comes directly from the  $1/k^2$  singularity of Eq. (7). The essential feature of the new term is that it produces damping of the spin-current proportional to the relative velocity between up- and down-spin electrons. This makes it readily distinguishable from the usual viscous friction contained in the second term, which is proportional to the *derivatives* of the velocity field. The physical reason for the difference is that, whenever up and down-spin currents travel with different average velocities, they exert *friction* on each other: the ‘‘spin drag coefficient’’ is  $\gamma(\omega) = [in^3A(\omega)/4\omega mn_\uparrow n_\downarrow]$ . Of course, like all the quantities considered here,  $\gamma(\omega)$  is complex and frequency dependent, and, in the limit of zero frequency, its real part can be shown to be related to the spin

$$\text{Im}A(\omega) \simeq -\frac{4}{3n^2V}\sum_{\vec{q}}v_{\vec{q}}^2q^2\int_0^\omega\frac{d\omega'}{\pi}[\text{Im}\chi_{\uparrow\uparrow}(q,\omega-\omega')\text{Im}\chi_{\uparrow\downarrow}(q,\omega') - \text{Im}\chi_{\uparrow\downarrow}(q,\omega-\omega')\text{Im}\chi_{\uparrow\uparrow}(q,\omega')], \quad (14)$$

which is exact in the limits of high density and high frequency. Here  $v_{\vec{q}} = \frac{4\pi e^2}{q^2}$  and  $\chi_{\sigma\sigma'}(q,\omega)$  are the spin-density response functions of the homogeneous liquid.

We have evaluated  $\chi_{\sigma\sigma'}$  in the generalized random phase approximation

$$\chi_{\sigma\sigma'}^{-1}(q,\omega) = [\chi_{\sigma\sigma'}^{(0)}]^{-1}(q,\omega)\delta_{\sigma\sigma'} - v_{\vec{q}}[1 - G_{\sigma\sigma'}(q)], \quad (15)$$

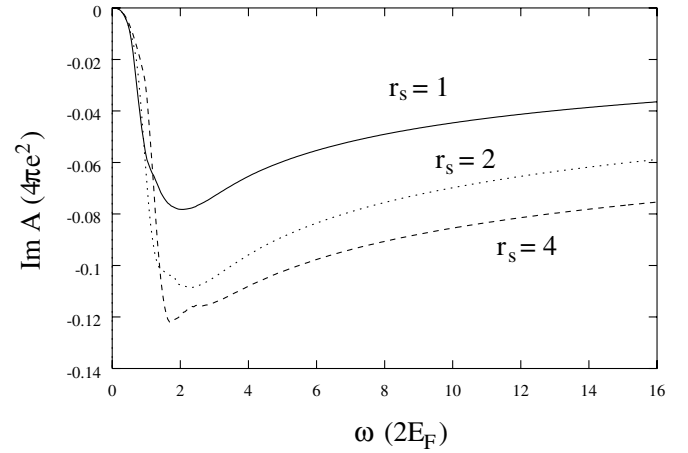


FIG. 1. Imaginary part of  $A(\omega)$  evaluated from Eq. (14) with the correction factor given in Eq. (16). The values of  $a(r_s, 0)$  are 1.92, 3.36, and 7.49 at  $r_s = 1, 2,$  and  $4,$  respectively.

diffusion constant  $D_s$  by the Einstein relation  $D_s = [n/m\chi_s\gamma(0)]$ , where  $\chi_s$  is the static, macroscopic spin susceptibility.

Unfortunately, an exact calculation of  $A(\omega)$  from the microscopic expressions (8) and (9) is beyond the reach of present-day many-body techniques. However, we can obtain a rather good approximation with the help of the following exact results: (i) For  $\omega \rightarrow 0$ ,  $\text{Im}A(\omega) \propto \omega^3$  and  $\text{Re}A(\omega) \propto \omega^2$ ; (ii) For large  $\omega$ ,  $\text{Im}A(\omega) \rightarrow -(16\pi e^2/3) \times (n_\uparrow n_\downarrow/n^2)(\alpha r_s/\sqrt{\bar{\omega}})[1/(1+\zeta)^{1/3}]$  and  $\text{Re}A(\omega) \rightarrow -(16\pi e^2/3)(n_\uparrow n_\downarrow/n^2)[g_{\uparrow\downarrow}(0) - 1]$ . Here  $\bar{\omega} = (\omega/2E_{F\uparrow})$ , where  $E_{F\uparrow}$  is the Fermi energy for majority spin electrons and  $\zeta = (n_\uparrow - n_\downarrow)/n$  measures the degree of spin polarization, and  $\alpha = (4/9\pi)^{1/3}$  [21]. Note that  $g_{\uparrow\downarrow}(0)$  is accurately known from the work of Gori-Giorgi and Perdew [22]. The high and low frequency limits of  $\text{Re}A(\omega)$  are both obtained from the third-moment sum rule. In particular, the vanishing of  $\text{Re}A(0)$  follows from the fact that  $\frac{2}{\pi}\int_0^\infty[\text{Im}A(\omega')/\omega']$  is equal to (minus) the first moment of the current-current response function, which, by gauge invariance and the continuity equation, coincides with the third-moment of the density-density response function, i.e.,  $-A(\infty)$ .

The  $\omega^3$  behavior of  $\text{Im}A(\omega)$  at low frequency is easily obtained from the approximate zero-temperature formula [17]

where  $\chi_{\sigma\sigma'}^{(0)}(q,\omega)$  is the Lindhard function and  $G_{\sigma\sigma'}(q)$  are local field corrections [23]. At typical metallic densities we multiply  $\text{Im}A(\omega)$  by an empirical factor

$$g(\omega) = \frac{1 + \sqrt{\bar{\omega}}}{a(r_s, \zeta) + \sqrt{\bar{\omega}}}, \quad (16)$$

designed to satisfy the condition  $\text{Re}A(0) = 0$  without

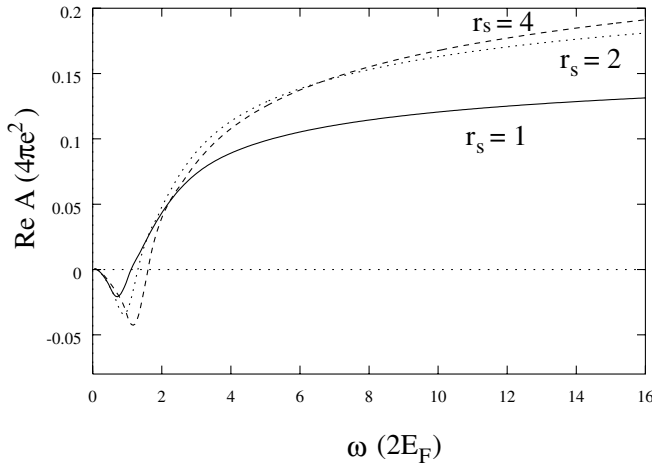


FIG. 2. Real part of  $A(\omega)$  obtained from Eq. (9).

altering the high-frequency behavior. Notice that  $a(r_s, \zeta) \rightarrow 1$  for  $r_s \rightarrow 0$ . The results evaluated with this procedure are shown in Figs. 1 and 2.

Finally, we briefly remark on the calculation of the regular part of the xc kernel. The spin symmetric combinations  $B_+^\alpha \equiv B_{\uparrow\uparrow}^\alpha + B_{\downarrow\downarrow}^\alpha$  with  $\alpha = L$  or  $T$  have been approximately calculated, in the paramagnetic state ( $\zeta = 0$ ), by two of us [24]. For the antisymmetric combinations  $B_-^\alpha \equiv B_{\uparrow\downarrow}^\alpha - B_{\downarrow\uparrow}^\alpha$  a provisional solution is offered by the time-honored Gross-Kohn interpolation formula [9], with parameters suitably tuned to satisfy the exact identity  $\lim_{\omega \rightarrow 0} \{ [B_{\sigma\sigma'}^L(\omega) - \frac{4}{3}B_{\sigma\sigma'}^T(\omega) - \epsilon_{xc,\sigma\sigma'}'' ] / \omega \} = 0$  [25], the third-moment sum rule, and the limiting form  $B_-^\alpha(\omega) \rightarrow \omega^{-3/2}$  for  $\omega \rightarrow \infty$ . Finally, for general polarization, we propose to use the formula  $B_{\sigma\sigma'}^\alpha(\zeta) = P_{\sigma\sigma'}(\zeta)B_{\sigma\sigma'}^\alpha(\zeta = 0)$  where  $P_{\sigma\sigma'}(\zeta) = [\epsilon_{xc,\sigma\sigma'}''(\zeta) / \epsilon_{xc,\sigma\sigma'}''(0)]$ , so that the above conditions are satisfied for all  $\zeta$ . This completes the construction of the input for Eq. (10). We hope that the new expression for the spin-current dependent xc field will stimulate applications of CDFT to the calculation of spin excitations in spin-polarized systems.

This work was supported by NSF Grant No. DMR-0074959. We acknowledge useful discussions with Carsten Ullrich, Paul de Boeij, and Robert van Leeuwen.

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