Surface Polariton Mode Densities

W. Eckhardt
Abt. Mathematische Physik, Universität Ulm

Z. Naturforsch. 38a, 10–15 (1983); received September 10, 1982

Starting from the Green functions of the electromagnetic (EM) field in two adjacent dielectric half-spaces, the surface and bulk contributions to the EM-density of states are discussed. The mode densities determine the thermodynamic properties of the radiation field.

1. Introduction

Localized solutions of the Maxwell equations on both sides of adjacent dielectric media are usually called surface polaritons. In the last decade in both – experimental and theoretical – studies the properties of these localized excitations were studied (see e.g. [1, 2] and references therein): dispersion relations, mechanism of excitation [3, 4, 5], research by Raman scattering [6, 7] etc.

Usually two different theoretical approaches are distinguished:

(i) Localized solutions of the Maxwell equations are directly constructed [1, 2];
(ii) The EM-boundary value problem is solved for the considered geometrical arrangement i.e., the Green functions are calculated (for this approach see e.g. [8, 9]).

Via the second method not only the surface polaritons are found but also all other contributions of the EM excitation: bulk polaritons, evanescent waves and oscillating parts (for a classification see e.g. [10]). The poles (resp. peaks if dissipation is not negligible) in the Green functions define the dispersion relations of the bulk and surface polaritons.

In this paper we are only interested in the thermodynamic properties of the EM-field in two adjacent dielectric half-spaces: the half-spaces \( z < 0 \) and \( z > 0 \) are characterized by \( \varepsilon_1(\omega) \) and \( \varepsilon_2(\omega) \), respectively. Plasma excitation and spatial dispersion are excluded.

We will calculate the Green functions and, using the fluctuation dissipation theorem (FDT) we will find the complete \( r \)-dependent density of states \( D(\omega, z) \) of the thermal EM-field.

The formal procedure corresponds to the method which was used in [11, 12]. The densities of states for both half-spaces consist of bulk, surface and diffuse contributions and determine the thermodynamic properties of the system.

2. Density of States

In lossless media with dispersion the local EM-energy can be expressed by the imaginary parts of the traces of the electric and magnetic Green functions (FDT) [11]:

\[
E(r, r, T) = \int_0^\infty \frac{d\omega}{2\pi} D(r, \omega) \frac{\hbar \omega}{2} \coth \frac{\beta \hbar \omega}{2}
\]

with

\[
D(r, \omega) = \frac{1}{4\pi^2 \omega} \text{Im} \left\{ \frac{\partial (\omega \varepsilon(r, \omega))}{\partial \omega} \left[ G^{\text{EE}}(r, r, \omega) + G^{\text{HH}}(r, r, \omega) \right] \right\}. \tag{2.2}
\]

The \( r \)-dependence is restricted to the separation of different homogeneous parts by sharp boundaries. In the considered geometry all fields can be expanded in terms of two dimensional plane waves in the \( x - y \) plane:

\[
D(z, \omega) = \frac{1}{4\pi^2 \omega} \int_\mathbb{R} \frac{d^2q}{(2\pi)^2} \text{Im} \left\{ \frac{\partial (\omega \varepsilon(z, \omega))}{\partial \omega} \left[ G^{\text{EE}}(q, z, z, \omega) + G^{\text{HH}}(q, z, z, \omega) \right] \right\}. \tag{2.3}
\]

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The calculation of the Green functions by standard methods yields (see e.g. [11, 13]):

(i) in the range $\Box, \text{i.e. } z, z' < 0$:

$$G_{\Box}^{\text{EE}}(q, z, z', \omega) = 2\pi i \frac{\omega^2}{c^2 k_{z1}} \left[ 2 \exp \left| ik_{z1}|z - z'| \right| \right. $$

$$+ \left[ \eta_{\perp} - \eta_{\parallel} \left( 1 - \frac{2q^2c^2}{\omega^2 \epsilon_1} \right) \right] \exp \left[ -i k_{z1}(z + z') \right] \left[ - \frac{4\pi}{\epsilon_1} \delta(z - z') \right], \tag{2.4}$$

$$G_{\Box}^{\text{HH}}(q, z, z', \omega) = 2\pi i \frac{\omega^2 \epsilon_1}{c^2 k_{z1}} \left[ 2 \exp \left| ik_{z1}|z - z'| \right| \right. $$

$$+ \left[ \eta_{\parallel} - \eta_{\perp} \left( 1 - \frac{2q^2c^2}{\omega^2 \epsilon_1} \right) \right] \exp \left[ -i k_{z1}(z + z') \right] \left[ - \frac{4\pi}{\epsilon_1} \delta(z - z') \right]. \tag{2.5}$$

(ii) in the range $\Box, \text{i.e. } z, z' > 0$:

$$G_{\Box}^{\text{EE}}(q, z, z', \omega) = 2\pi i \frac{\omega^2}{c^2 k_{z2}} \left[ 2 \exp \left| ik_{z2}|z - z'| \right| \right. $$

$$+ \left[ \eta_{\parallel} - \eta_{\perp} \left( 1 - \frac{2q^2c^2}{\omega^2 \epsilon_2} \right) \right] \exp \left[ -i k_{z2}(z + z') \right] \left[ - \frac{4\pi}{\epsilon_2} \delta(z - z') \right], \tag{2.6}$$

$$G_{\Box}^{\text{HH}}(q, z, z', \omega) = 2\pi i \frac{\omega^2 \epsilon_2}{c^2 k_{z2}} \left[ 2 \exp \left| ik_{z2}|z - z'| \right| \right. $$

$$+ \left[ \eta_{\perp} - \eta_{\parallel} \left( 1 - \frac{2q^2c^2}{\omega^2 \epsilon_2} \right) \right] \exp \left[ -i k_{z2}(z + z') \right] \left[ - \frac{4\pi}{\epsilon_2} \delta(z - z') \right]. \tag{2.7}$$

In (2.4) – (2.7) we defined the quantities:

$$k_{z1} = \left( \frac{\omega^2}{c^2} \epsilon_1 - q^2 \right)^{1/2}, \tag{2.8}$$

$$k_{z2} = \left( \frac{\omega^2}{c^2} \epsilon_2 - q^2 \right)^{1/2}, \tag{2.9}$$

$$\eta_{\parallel} = (\epsilon_2 k_{z1} - \epsilon_1 k_{z2}) (\epsilon_2 k_{z1} + \epsilon_1 k_{z2})^{-1}, \tag{2.10}$$

$$\eta_{\perp} = (k_{z1} - k_{z2}) (k_{z1} + k_{z2})^{-1}. \tag{2.11}$$

The translationally invariant parts of (2.4) – (2.7) are easily evaluated:

$$D_{\Box}^{\text{EE}}(\omega) = \theta(\epsilon_1) \frac{\omega^2}{\pi^2 c^3} \frac{d(\omega \sqrt{\epsilon_1})}{d\omega}, \tag{2.12}$$

$$D_{\Box}^{\text{HH}}(\omega) = \theta(\epsilon_2) \frac{\omega^2}{\pi^2 c^3} \frac{d(\omega \sqrt{\epsilon_2})}{d\omega}. \tag{2.13}$$

$\theta$ denotes the Heaviside step function.

Equations (2.12) and (2.13) may be interpreted as the density of states of the bulk polaritons $^*$. 

$^*$ Due to the expansion in terms of plane waves in the $x - y$ plane the poles in the translationally invariant parts of (2.4) – (2.7) do not represent the dispersion relation of bulk polaritons. This is contrary to the expansion in terms of 3-dim. plane waves (spatial Fourier transformation) for the isotropic infinite space. The identification of (2.12) and (2.13) as bulk contributions becomes obvious if we note that in (2.12) and (2.13) all translationally invariant parts with real $k_{z1}$ i.e. $(\omega^2/c^2) \epsilon_1 \geq q^2$ are included ($\epsilon_i > 0$).
In the following we assume that there is no dispersion in the half-space \( z > 0 \) (\( \varepsilon_2 = \text{const} > 0 \)). We immediately note that the imaginary parts of \( \eta_1 \) and \( \eta_\perp \) vanish for \( q < \min((\omega/c) \sqrt{\varepsilon_1}) \) or \( q > \max((\omega/c) \sqrt{\varepsilon_1}) \) if \( \varepsilon_1 > 0 \) and for \( q > (\omega/c) \sqrt{\varepsilon_2} \) if \( \varepsilon_1 < 0 \).

Furthermore we note that for \( \varepsilon_1 < -\varepsilon_2 \) there is a pole in \( \text{Im} \eta_1 \) at \( q^2 = \frac{\omega^2}{c^2} \frac{\varepsilon_1}{|\varepsilon_1 - \varepsilon_2|} \) (i.e. \( q > (\omega/c) \sqrt{\varepsilon_2} \)). The last formula represents the dispersion relation of surface polaritons.

The contributions due to the pole in \( \eta_1 \) are taken into account by a limiting process: At first we assume \( \varepsilon_2 = 0 \) and then we take the limit \( \varepsilon_2 \to 0 \). This procedure guarantees causality resp. the appropriate Kramers-Kronig relations for lossless media.

Introducing the abbreviations

\[
A(\omega, q) = \frac{1}{\varepsilon_1} \frac{d(\omega \varepsilon_1)}{d\omega} \left[ \eta_\perp - \eta_1 \left( 1 - \frac{2q^2}{\omega^2 \varepsilon_1} \right) \right] + \left[ \eta_1 - \eta_\perp \left( 1 - \frac{2q^2}{\omega^2 \varepsilon_1} \right) \right]
\]

and

\[
b(\omega, q) = \eta_1 + \eta_\perp,
\]

we can write the complete corrections to the bulk contributions (2.12) and (2.13) in the form:

\[
\Delta D^1(\omega, z) = \theta(\varepsilon_1) \left\{ \frac{\omega \varepsilon_1}{4\pi^2 c^2} \frac{q dq}{k_\perp^2} \text{Re} A(\omega, q) \cos 2k_\perp z \right\}
\]

\[
- \frac{\omega \varepsilon_1}{4\pi^2 c^2} \theta(\varepsilon_1 - \varepsilon_2) \int \frac{q dq}{(\omega/c) \sqrt{\varepsilon_2} k_\perp} \text{Im} A(\omega, q) \sin 2k_\perp z
\]

\[
+ \frac{\omega \varepsilon_1}{4\pi^2 c^2} \theta(\varepsilon_2 - \varepsilon_1) \int \frac{q dq}{(\omega/c) \sqrt{\varepsilon_1} k_\perp} \text{Im} A(\omega, q) \exp \{ 2k_\perp z \}
\]

\[
+ \theta(-\varepsilon_1) \left\{ \frac{\omega \varepsilon_1}{4\pi^2 c^2} \frac{q dq}{k_\perp^2} \text{Im} A(\omega, q) \exp \{ 2k_\perp z \} \right\}
\]

\[
+ \frac{\omega}{4\pi^2 c^2} \theta(-\varepsilon_2 - \varepsilon_1) \int \frac{q dq}{(\omega/c) \sqrt{\varepsilon_1} k_\perp} \left[ - \frac{d(\omega \varepsilon_1)}{d\omega} \left( 1 - \frac{2q^2}{\omega^2 \varepsilon_1} \right) + \varepsilon_1 \right]
\]

\[
\times \lim_{\varepsilon_2 \to 0} \text{Im} \eta_1 \exp \{ 2k_\perp z \}
\]

\[
\Delta D^2(\omega, z) = \theta(\varepsilon_1) \left\{ -\frac{1}{2\pi^2 \omega} \int \frac{q^3 dq}{k_\perp^2} \text{Re} B(\omega, q) \cos 2k_\perp z \right\}
\]

\[
+ \frac{1}{2\pi^2 \omega} \theta(\varepsilon_2 - \varepsilon_1) \int \frac{q^3 dq}{(\omega/c) \sqrt{\varepsilon_1} k_\perp} \text{Im} B(\omega, q) \sin 2k_\perp z
\]

\[
+ \frac{1}{2\pi^2 \omega} \theta(\varepsilon_1 - \varepsilon_2) \int \frac{q^3 dq}{(\omega/c) \sqrt{\varepsilon_2} k_\perp} \text{Im} B(\omega, q) \exp \{ -2k_\perp z \}
\]

\[
+ \theta(-\varepsilon_1) \left\{ -\frac{1}{2\pi^2 \omega} \int \frac{q^3 dq}{k_\perp^2} \left[ \text{Re} B(\omega, q) \cos 2k_\perp z - \text{Im} B(\omega, q) \sin 2k_\perp z \right] \right\}
\]

\[
- \frac{1}{2\pi^2 \omega} \theta(-\varepsilon_2 - \varepsilon_1) \int \frac{q^3 dq}{(\omega/c) \sqrt{\varepsilon_1} k_\perp} \lim_{\varepsilon_2 \to 0} \text{Im} \eta_1 \exp \{ -2k_\perp z \}
\]

\[
\]
The boundary conditions demand the continuity of $q$ at the boundary. Therefore, we may indentify the “diffuse” and “sharp” modes which correspond to each other on both sides of the boundary.

For $\epsilon_1 > 0$ and $\epsilon_1 > \epsilon_2$ we find in region $\Omega$ only oscillating cosinus contributions for $q < (\omega/c) \sqrt{\epsilon_2}$. The corresponding modes in region $\Omega$ show the same behaviour.

For $(\omega/c) \sqrt{\epsilon_1} < q < (\omega/c) \sqrt{\epsilon_2}$ we observe in region $\Omega$ oscillating cosinus and sinus contributions. The corresponding modes in region $\Omega$ are spatially damped (evanescent) waves. In the optically denser medium there are no damped contributions.

For $\epsilon_1 < \epsilon_2$ the analogous considerations are valid. For $\epsilon_1 < 0$ (we assumed $\epsilon_2 > 0$) there are only damped contributions in medium $\Omega$ with $q < (\omega/c) \sqrt{\epsilon_2}$. The corresponding modes in medium $\Omega$ are oscillating cosinus and sinus contributions.

If additionally $\epsilon_1 < -\epsilon_2$ there are “sharp” localized modes on both sides of the boundary: surface polaritons. Usually ($\epsilon_1 < -\epsilon_2$) the oscillating and damped (evanescent) contributions are denoted as diffuse parts of the surface polaritons [9, 10].

Generally, the diffuse parts may be neglected compared with the surface polaritons [9].

Summing up (2.12), (2.13), (2.16) and (2.17) we can write the complete density of states in the compact formula:

$$D^\Omega(\omega, z) = \theta(\epsilon_1) D^\Omega_{\Omega}(\omega) \left[ 1 + d_{\text{pec}}^\Omega + \theta(\epsilon_1 - \epsilon_2) d_{\text{pec}}^\Omega + \theta(\epsilon_2 - \epsilon_1) d_{\text{pec}}^\Omega \right]$$

$$+ \theta(-\epsilon_1) D^\Omega_{\Omega}(\omega) \left[ d_{\text{pec}}^\Omega + \theta(-\epsilon_2 - \epsilon_1) d_{\text{pol}}^\Omega \right]$$

$$D^\Omega_{\Omega}(\omega) = \frac{\omega^2 \epsilon_1 \sqrt{\epsilon_1}}{\pi^2 c^3}.$$  

(2.18)

Introducing the proper substitutions in the integrals of (2.16) and (2.17) we can write the correction parts of (2.18) and (2.19) in the form:

$$d_{\text{pec}}^\Omega = \frac{1}{4} \sqrt{\epsilon_1} \left( \frac{d(\omega \sqrt{\epsilon_1})}{d\omega} \right)^{-1} \int_0^1 dy_1 \operatorname{Re} A(\omega, q(y_1)) \cos \beta_1 y_1,$$

$$d_{\text{pol}}^\Omega = \frac{1}{4} \sqrt{\epsilon_1} \left( \frac{d(\omega \sqrt{\epsilon_1})}{d\omega} \right)^{-1} \int_0^1 dy_1 \operatorname{Im} A(\omega, q(y_1)) \sin \beta_1 y_1,$$

(2.21)

(2.22)

where

$$y_1 = \left[ 1 - \frac{q^2 c^2}{\omega^2 \epsilon_1} \right]^{1/2}.$$  

(2.23)

$$d_{\text{pec}}^\Omega = \frac{1}{4} \sqrt{\epsilon_1} \left( \frac{d(\omega \sqrt{\epsilon_1})}{d\omega} \right)^{-1} \int_0^1 dy_1 \operatorname{Im} A(\omega, q(y_1)) \exp \{ \beta_1 y_2 \},$$

(2.24)

where

$$y_2 = \left[ \frac{q^2 c^2}{\omega^2 \epsilon_1} - 1 \right]^{1/2},$$  

(2.25)

$$d_{\text{pec}}^\Omega = \frac{1}{4} \sqrt{\epsilon_1} \int_{1}^{\infty} dy_3 \operatorname{Im} A(\omega, q(y_3)) \exp \{ \beta_1 y_3 \},$$

(2.26)

$$d_{\text{pol}}^\Omega = \frac{1}{4} \sqrt{\epsilon_1} \int_{1}^{\infty} dy_3 \left[ \frac{1}{\epsilon_1} \frac{d(\omega \epsilon_1)}{d\omega} (1 + y_3^2) + 1 \right] \lim_{\epsilon_2 \to 0} \operatorname{Im} \eta_1(\omega, q(y_3)) \exp \{ \beta_1 y_3 \},$$  

(2.27)
where
\[ y_3 = \left[ 1 + \frac{q^2 c^2}{\omega^2 |e_1|} \right]^{1/2}. \]  

In medium 2 we find:
\[ g_3^{\text{pec}} = -\frac{1}{2} \int_0^1 (1 - y_3^2) \, dy_4 \Re B(\omega, q(y_4)) \cos \beta_2 y_4, \]  
\[ g_2^{\text{pec}} = \frac{1}{2} \int_0^{\sqrt{1 - \frac{q^2 c^2}{\omega^2 e_2}}} (1 - y_2^2) \, dy_4 \Im B(\omega, q(y_4)) \sin \beta_2 y_4, \]  
with \[ y_4 = \left[ 1 - \frac{q^2 c^2}{\omega^2 e_2} \right]^{1/2}. \]
\[ g_3^{\text{loc}} = \frac{1}{2} \int_0^{\sqrt{\frac{q^2 c^2}{\omega^2 e_2} - 1}} (1 + y_3^2) \, dy_4 \Im B(\omega, q(y_3)) \exp \{-\beta_2 y_3\}, \]  
with \[ y_5 = \left[ \frac{q^2 c^2}{\omega^2 e_2} - 1 \right]^{1/2}. \]
\[ g_3^{\text{loc}} = -\frac{1}{2} \int_0^1 (1 - y_3^2) \, dy_4 \Re B(\omega, q(y_4)) \cos \beta_2 y_4 - \Im B(\omega, q(y_4)) \sin \beta_2 y_4, \]  
\[ g_2^{\text{loc}} = -\frac{1}{2} \int_0^\infty (1 + y_3^2) \, dy_4 \lim_{\varepsilon_2 \to 0} \Im \eta(\omega, q(y_5)) \exp \{-\beta_2 y_3\}. \]  

In (2.21) – (2.35) we have defined the frequency scaled distances to the boundary:
\[ \beta_i = 2 \frac{\varepsilon_i}{c} \sqrt{|\varepsilon_i| \cdot z}, \quad i = 1, 2. \]  

In the limit \( z \to \pm \infty \) all corrections tend to zero as \( 1/z \). This can easily be seen by changing the integration variables suitably (\( \beta \) \( y \to t \)). (In the limit \( \beta \to \infty \) the coefficients \( A \) and \( B \) do not longer depend on the new variable \( t \).)

### 3. Surface Polariton Mode Density

The physically most important terms are represented by \( d_2^{\text{pol}} \) and \( g_3^{\text{pol}} \).

The limiting process \( \varepsilon_2^2 \to 0 \) yields (we omit the index of \( y \)):
\[ \lim_{\varepsilon_2^2 \to 0} \Im \eta(q(y)) = \lim_{\varepsilon_2^2 \to 0} \Im \frac{\varepsilon_2 y + |e_1|}{\varepsilon_2 y - |e_1|} \sqrt{y^2 - (1 + \varepsilon_2/y/e_2) y^2 - (1 + \varepsilon_2/y/e_2)} \]
\[ = -2 \sqrt{y^2 - (1 + \frac{\varepsilon_2}{|e_1|}) |e_1| \pi \delta \left( \frac{\varepsilon_2 y - |e_1|}{|e_1|} \sqrt{y^2 - (1 + \frac{\varepsilon_2}{|e_1|})} \right)} \].

Expanding the \( \delta \)-function we find:
\[ \delta \left( \frac{\varepsilon_2 y - |e_1|}{|e_1|} \sqrt{y^2 - (1 + \frac{\varepsilon_2}{|e_1|})} \right) = \frac{\varepsilon_2}{|e_1|^2 - \varepsilon_2^2} \delta \left( y - \sqrt{\frac{|e_1|}{|e_1| - e_2}} \right). \]

The pole of the \( \delta \)-function lies in the considered integration range and we obtain:
\[ d_2^{\text{pol}} = \frac{1}{4} \frac{1}{|e_1|} \frac{d(e_2/|e_1|)}{d\varepsilon_1} \left( 1 + \frac{|e_1|}{|e_1| - e_2} \right) + \frac{\varepsilon_2}{|e_1|^2 - \varepsilon_2^2} \exp \left( \beta_1 \sqrt{\frac{|e_1|}{|e_1| - e_2}} \right). \]
After some trivial manipulations we find the $z$-dependent density of states of the surface polaritons:

$$D_{\text{pol}}^0(\omega, z) = \theta(-\epsilon_1) D_0^0 \theta(-\epsilon_2 - \epsilon_1) D_{\text{pol}}^2 = \theta(-\epsilon_1 - \epsilon_2) \frac{\omega^2}{\pi c^3} \frac{\epsilon_1 \epsilon_2}{(\epsilon_1 + \epsilon_2)(\epsilon_1 - \epsilon_2)^{5/2}}$$

$$\cdot \left\{ \omega \frac{d\epsilon_1}{d\omega} \left| \frac{\epsilon_1 + \epsilon_2}{2} \right| - \left| \frac{\epsilon_1}{\epsilon_2} \right| \right\} \exp \left( \frac{2\omega}{c} \sqrt{\frac{\epsilon_1^2}{\epsilon_1 - \epsilon_2}} z \right). \quad (3.4)$$

The validity of the fundamental inequalities $[14] d(\omega \epsilon)/d\omega > 0$ and $\omega d\epsilon/d\omega > 2(1 - \epsilon)$ guarantees the positivity of (3.4).

Completely analogous calculations lead to $\tilde{g}_{\text{pol}}^2$; we find:

$$D_{\text{pol}}^2(\omega, z) = \theta(-\epsilon_1) D_0^2(\omega) \theta(-\epsilon_2 - \epsilon_1) D_{\text{pol}}^2$$

$$= \theta(-\epsilon_1 - \epsilon_2) \frac{\omega^2 \epsilon_2^3}{\pi c^3} \frac{|\epsilon_1|^3}{(\epsilon_1 + \epsilon_2)(\epsilon_1 - \epsilon_2)^{5/2}} \exp \left\{ - \frac{2\omega}{c} \sqrt{\frac{\epsilon_2^2}{\epsilon_1 - \epsilon_2}} z \right\}. \quad (3.5)$$

(for $\epsilon_2 = 1$ this expression has already been given in [11]).

Integration over $z$ yields the total polariton energy per unit area in the $x - y$ plane:

$$\frac{E_{\text{pol}}}{S} = \int_0^\infty \omega \hbar \omega \left( \frac{1}{2} + \left[ e^{\hbar\omega/k_B T} - 1 \right]^{-1} \right) \theta(-\epsilon_1 - \epsilon_2) \frac{\omega}{2 \pi c^2} \frac{\epsilon_2}{(\epsilon_1 + \epsilon_2)(\epsilon_1 - \epsilon_2)^2}$$

$$\cdot \left[ \omega \frac{d\epsilon_1}{d\omega} \frac{\epsilon_1 + \epsilon_2}{2} - \epsilon_1 \left( \frac{\epsilon_1^2}{\epsilon_1 - \epsilon_2} \right) \right]. \quad (3.6)$$

### 4. Conclusions

In this paper the EM-excitations of a dielectric boundary layer were discussed. For the description of these excitations we defined a local density of states including all bulk and surface contributions on both sides of the boundary: the bulk polaritons, the sharp and diffuse surface polariton contributions and the oscillating and damped (evanescent) waves.

The dominant surface contribution of the density of states in the frequency range for which $\epsilon_1 < -\epsilon_2$ is given by the sharp surface polaritons. This contribution was used to derive the internal energy of the surface polaritons (3.6).

In concrete cases, when $\epsilon_1$ and $\epsilon_2$ are known, this formula allows us to calculate the thermodynamic properties of these elementary excitations.