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Laser Excitation of Polarization Waves in a Frozen Gas

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Abstract

Laser experiments with optically excited frozen gases entail the excitation of polarization waves. In a continuum approximation the waves are dispersionless, but their frequency depends on the direction of the propagation vector. An outline is given of the theory of transient phenomena that involve the excitation of these waves by a resonant dipole-dipole transfer process.

1. INTRODUCTION

If the energy associated with the translational temperature of a gas is low compared to the relevant excitation energies and to the energy associated with the inverse observation time, then the gas can be considered as "frozen" and is analogous to an amorphous solid, or a glass. Such a frozen gas can be produced by laser cooling, and the atoms or molecules can be selectively placed in certain excited states. When only a few states are involved, the system is analogous to a spin glass and can support polarization waves (dipolar Frenkel excitons) that are analogous to spin waves. In this paper, we introduce a basic example of such waves and discuss their excitation.

In laser experiments, the frozen gas is set up in a non-equilibrium electronic state, and the evolution of this state is measured after some elapsed time. Relaxation processes can be neglected for the duration of the experiment. Thus, the electronic state evolves towards dynamical equilibrium through the excitation of polarization waves in the random medium of the frozen gas. The theory of this process must determine the spectrum of the polarization waves as well as their excitation probability during the transient evolution to dynamical equilibrium. The two aspects of the problem will often be intermingled. However, we have chosen to work on a system where they can be separated to some extent.

The system under consideration is fully described in an earlier paper by J. S. Frasier, V. Celli, and T. Blum,[1] henceforth to be referred to as FCB. In the present paper, we develop an improved theory of the particular case that is discussed in Sec. III of FCB, under the heading "Sparse $ss' \to pp'$ in a bath of $sp \to ps$." Simply put, we consider one atom initially in state s' that is immersed in a frozen gas of atoms in state s. The s' state is connected to a p' state by a dipole matrix element μ', and an s state is connected to a p state by a dipole matrix element μ. The p and p' states are singled out from all possible states, because the $ss' \to pp'$ transition is resonant, i.e., $\epsilon'_s + \epsilon_s = \epsilon'_p + \epsilon_p$, and it is known that the transition rate peaks sharply at resonance.[2,3] Deviations from resonance are not considered in this paper. The resonance condition is achieved by the application of a static electric field. The $ss' \to pp'$ process excites polarization waves described by $sp \to ps$ transitions in the gas of s atoms. Typically, the number N of s atoms in the volume Ω is such that $N/\Omega \simeq 10^9$ atoms/cm^3. The s' atoms are "sparse" if their number N' is such that $N'/N \ll 1$, and in that case the result for N' atoms can be obtained by computing the result for one s' atom, averaging over distributions of the s atoms, and then multiplying this result by N'.

2. THE EQUATIONS OF TIME EVOLUTION

We begin with the set of coupled differential equations in Eq. (24) of FCB, Fourier transformed in time:

$$\omega a_0 = \sum_l V_l c_l + i, \tag{1a}$$

$$\omega c_l = V_l a_0 + \sum_m U_{lm} c_m. \tag{1b}$$

As in FCB, $a_0(t)$ is the amplitude of the state in which the atom at the origin is in state s' and all other atoms are in state s, while $c_l(t)$ is the amplitude of the state in which the atom at the origin is in state p', the atom at position $\mathbf{r}_l$ is in state p, and all other atoms are in state s. The quantity V_l is the interaction potential

$$V(\mathbf{r}) = \frac{\mu\mu'}{r^3}\left(1 - 3\cos^2\theta_r\right) = -2\frac{\mu\mu'}{r^3}P_2(\cos\theta_r), \tag{2}$$

evaluated at $\mathbf{r} = \mathbf{r}_l$, and θ_r is the polar angle. Similarly, U_{lm} is

$$U(\mathbf{r}) = \frac{\mu^2}{r^3}\left(1 - 3\cos^2\theta_r\right) = -2\frac{\mu^2}{r^3}P_2(\cos\theta_r), \tag{3}$$

evaluated at $\mathbf{r} = \mathbf{r}_l - \mathbf{r}_m$. The angular factor present in V and U has been chosen for mathematical convenience: the actual angular factor is more complicated, as discussed in Sec. 5. In FCB, all the results depend only on an angular average of V^2 and, as is shown in the Appendix of that paper, one can work with the effective angle-averaged potential $V(r) = \left(4/3\sqrt{3}\right)\mu\mu'/r^3$. In the treatment presented here, however, the angular dependence plays a more important role.

The potentials of Eqs. (2) and (3) have particularly simple Fourier transforms. Defining

$$V(\mathbf{k}) = \int d^3r\, V(\mathbf{r})\, e^{-i\mathbf{k}\cdot\mathbf{r}}, \tag{4}$$

we find, using the standard expansion of $e^{-i\mathbf{k}\cdot\mathbf{r}}$ in spherical Bessel functions and spherical harmonics,[4]

$$\begin{aligned} V(\mathbf{k}) &= -8\pi\mu\mu' P_2(\cos\theta_k) \int_0^\infty \frac{dr}{r} j_2(kr) \\ &= -\frac{8\pi}{3}\mu\mu' P_2(\cos\theta_k). \end{aligned} \tag{5}$$

Similarly,

$$U(\mathbf{k}) = -\frac{8\pi}{3}\mu^2 P_2(\cos\theta_k)\,. \tag{6}$$

With these expressions we can obtain explicit solutions for the polarization waves in the continuum approximation, and we can then use these approximate solutions to describe the dynamics of a frozen gas state.

3. THE CONTINUUM MODEL

We change the sums in Eqs. (1) to integrations over space, so that the evolution equations become

$$\omega a_0 = \frac{N}{\Omega}\int d^3r\, V(\mathbf{r})\, c(\mathbf{r}) + i, \tag{7a}$$

$$\omega c(\mathbf{r}) = V(\mathbf{r})\, a_0 + \frac{N}{\Omega}\int d^3r'\, U(\mathbf{r}-\mathbf{r}')\, c(\mathbf{r}')\,, \tag{7b}$$

where $c(\mathbf{r})$ is now an averaged quantity. Fourier transforming, we obtain simply

$$\omega a_0 = \frac{N}{\Omega}\int \frac{d^3k}{(2\pi)^3}\, V(\mathbf{k})\, c(\mathbf{k}) + i \tag{8a}$$

$$\omega c(\mathbf{k}) = V(\mathbf{k})\, a_0 + \frac{N}{\Omega} U(\mathbf{k})\, c(\mathbf{k})\,. \tag{8b}$$

The last equation shows that the frequencies of the polarization waves are

$$\omega(\mathbf{k}) = \frac{N}{\Omega} U(\mathbf{k})\,, \tag{9}$$

independent of the magnitude of $\mathbf{k}$, but dependent on the direction, according to Eq. (6). This peculiar dispersion relation, which is analogous to well-known results in spin wave theory, is a consequence of the long range nature of the dipole-dipole interaction. We expect it to be useful for values of k up to the order of the inverse of the interatomic spacing.[5] The solution of Eqs. (8) gives

$$a_0(\omega) = \frac{i}{\omega - M(\omega)}, \tag{10}$$

where

$$M(\omega) = \frac{N}{\Omega}\int \frac{d^3k}{(2\pi)^3}\frac{V(\mathbf{k})^2}{\omega - \omega(\mathbf{k})}. \tag{11}$$

Carrying out the angular integration, we find

$$M(\omega) = -\frac{4}{3\pi}(\mu')^2\left[\int_0^{k_c} dk\, k^2\right] m(z)\,, \tag{12}$$

where, leaving for Sec. 4 a discussion of the effective cutoff k_c,

$$m(z) = z + \frac{z^2}{\sqrt{3+6z}} \log\left(\frac{1-\sqrt{\frac{3}{1+2z}}}{1+\sqrt{\frac{3}{1+2z}}}\right), \tag{13}$$

and $z = \omega/u_0$ with

$$u_0 = \frac{8\pi}{3}\mu^2\frac{N}{\Omega}. \tag{14}$$

A plot of $\mathrm{Re}\, m$ and $\mathrm{Im}\, m$ on the real axis is shown in Fig. 1. For large z,

$$m(z) \sim -\frac{1}{5z} + O\left(\frac{1}{z^2}\right). \tag{15}$$

As expected, $m(z)$ is an analytic function in the upper z-plane, with a cut extending from $z = -1/2$ to $z = 1$. On the real axis outside the cut $m(z)$ is real, and the spectral density on the cut is given by

$$\mathrm{Im}\left[m(z)\right] = \frac{\pi}{\sqrt{6}}\frac{z^2}{\sqrt{z+\frac{1}{2}}}. \tag{16}$$

4. CONTINUUM APPROXIMATION FOR THE RANDOM MEDIUM

To complete the evaluation of the self-energy $M(\omega)$ of Eq. (12), it is necessary to introduce a cutoff in the k integral. We can do this in a way that makes the result applicable, at least approximately, to a random medium.

We know from Sec. II of FCB that the solution of Eqs. (1) for $U = 0$ is of the form of Eq. (10), with

$$M(\omega)_{U=0} = \frac{1}{\omega}\sum_l V_l^2, \tag{17}$$

while in the continuum limit, using Eq. (5),[6]

$$\begin{aligned} M(\omega)_{U=0} &= \frac{N}{\Omega}\int \frac{d^3k}{(2\pi)^3}\frac{V(\mathbf{k})^2}{\omega} \\ &= \frac{32}{45}\frac{N}{\Omega}\left[\int_0^{k_c} dk\, k^2\right]\frac{(\mu\mu')^2}{\omega}. \end{aligned} \tag{18}$$

Therefore, we see that if we make the substitution

$$\frac{N}{\Omega}\int_0^{k_c} dk\, k^2 \to \frac{45}{8}\sum_l \frac{1}{r_l^6}\left[P_2\left(\cos\theta_{r_l}\right)\right]^2, \tag{19}$$

then in the limit of U going to zero our continuum result will agree with the result of FCB. Applying this substitution, we find that Eq. (12) becomes

$$M(\omega) = -\frac{15}{2\pi}\frac{\Omega}{N}(\mu')^2 m(z)\sum_l \frac{1}{r_l^6}\left[P_2(\cos\theta_{r_l})\right]^2$$
$$= -\frac{5m(z)}{u_0}\sum_l V_l^2. \tag{20}$$

We can now average over atom positions by the technique described in FCB, Eq. (9), with the result that, for $\operatorname{Im} z > 0$,

$$(a_0)_{av} = \int_0^\infty d\beta \, \exp\left(i\beta\omega - v\sqrt{-\frac{5i\beta}{u_0}m(z)}\right)$$
$$= \int_0^\infty \frac{d\beta}{u_0}\exp\left(i\beta z - \frac{v}{u_0}\sqrt{-5i\beta m(z)}\right), \tag{21}$$

where, as in FCB Eq. (A2),

$$v = \frac{16\pi^{3/2}}{9\sqrt{3}}\frac{N}{\Omega}\mu\mu', \tag{22}$$

so that

$$\frac{v}{u_0} = \frac{2\sqrt{3\pi}}{9}\frac{\mu'}{\mu}. \tag{23}$$

The integral in Eq. (21) can be expressed in terms of the complementary error function.[1]

To develop approximations that automatically include a momentum cutoff we introduce the Green's function of Eq. (1b), which satisfies the equation

$$\omega G_{ln} = \delta_{ln} + \sum_m U_{lm}G_{mn}. \tag{24}$$

Then the solution of Eqs. (1) is of the form of Eq. (10), with the exact self-energy given by

$$M(\omega) = \sum_{lm} V_l G_{lm} V_m. \tag{25}$$

Similarly, the on-site Green's function, which clearly cannot be treated in the continuum approximation, can be written as

$$G_{nn} = \frac{1}{\omega - \sum_{mp} U_{nm}G_{mp,[n]}U_{pn}}, \tag{26}$$

while, for $l \neq n$,

$$G_{ln} = G_{ll,[n]}U_{ln}G_{nn} + \sum_{m\neq l} G_{lm,[n]}U_{mn}G_{nn}, \tag{27}$$

where $G_{lm,[n]}$ is the Green's function for a medium where the n atom is absent. These equations are exact, but we now replace all occurrences of G_{nn} on the right hand side by

$\langle G\rangle$, where $\langle G\rangle$ denotes the average value of G_{nn}, which does not depend on n. Averaging and Fourier transforming the last equation and setting

$$\langle G_{lm}\rangle = \int \frac{d^3k}{(2\pi)^3} G(\mathbf{k}) \exp\left[i\mathbf{k}\cdot(\mathbf{r}_l - \mathbf{r}_m)\right] \tag{28}$$

we obtain

$$\begin{aligned} G(\mathbf{k}) &= \frac{\langle G\rangle^2 U(\mathbf{k})}{1-(N/\Omega)\langle G\rangle U(\mathbf{k})} \\ &= \frac{\Omega}{N}\left(\frac{1}{\langle G\rangle^{-1}-(N/\Omega)U(\mathbf{k})} - \langle G\rangle\right). \end{aligned} \tag{29}$$

Similarly, writing the approximate version of Eq. (26) in the form

$$\begin{aligned} G_{nn} &= \frac{1}{i}\int_0^\infty d\beta \, \exp\left(i\beta\omega - i\beta\sum_m U_{nm}^2 \langle G\rangle\right) \\ &\times \exp\left(-i\beta\sum_{m\neq p} U_{nm} G_{mp,[n]} U_{pn}\right), \end{aligned} \tag{30}$$

and expanding the second exponential, we obtain the self-consistent equation

$$\begin{aligned} \langle G\rangle &= \frac{1}{i}\int_0^\infty d\beta \, \exp\left(i\beta\omega + (-i\beta\langle G\rangle)^{1/2} u\right) \\ &\times \left(1 - 2i\beta\int \frac{d^3k}{(2\pi)^3} U_{eff}(\mathbf{k})^2 G(\mathbf{k}) + \ldots\right), \end{aligned} \tag{31}$$

where $U_{eff}(\mathbf{k})$ is the Fourier transform of

$$U_{eff}(\mathbf{r}) = U(\mathbf{r}) \exp\left(-i\beta\langle G\rangle U^2(\mathbf{r})\right). \tag{32}$$

This shows how a cutoff is automatically introduced into the theory. With these values of $\langle G\rangle$ and $\langle G_{lm}\rangle$ we have approximately

$$M(\omega) = \sum_l V_l^2 \langle G\rangle + \sum_{l\neq n} V_l \langle G_{ln}\rangle V_n. \tag{33}$$

The self-consistency equation (32) has been discussed extensively in the approximation where the contribution of $G(\mathbf{k})$ is neglected (i.e., where contents of the last set of parentheses in Eq. (32) are replaced by unity).[7] It is believed that this approximation is adequate when the polarization waves are localized in the random medium.

If $\langle G\rangle$ is known as a function of ω, one can obtain $\langle a_0\rangle$ directly. Comparing Eq. (26) with

$$a_0(\omega) = \frac{i}{\omega - \sum_{lm} V_l G_{lm} V_m}, \tag{34}$$

we see that for an infinite medium

$$\langle a_0(\omega)\rangle = i\left(\frac{\mu}{\mu'}\right)^2 \left\langle G\left(\omega\left(\frac{\mu}{\mu'}\right)^2\right)\right\rangle . \tag{35}$$

It is also possible, in principle, to compute the average of $|a_0(t)|^2$ for the frozen gas. The result for $a_0(\omega)$ with the $M(\omega)$ of Eq. (20) can be inverse Fourier transformed and squared, and then the result must be averaged over all $\mathbf{r}$. We have already discussed the cut of $m(z)$ in Sec. 3, and one can see from Eq. (10) and the plots of Fig. 1 that $a_0(t)$ will have a pole at some $z < -1/2$, a pole at $z = 0$, and a pole at some $z > 1$. Therefore the inverse Fourier transform can be computed by summing the residues of these poles and integrating around the cut, a process which is amenable to numerical techniques. However, the result is unreliable because the pole at $z = 0$ is not really present in a random medium. (In the continuum approximation, waves of zero frequency travel in the direction where $P_2(\cos\theta) = 0$ and are not excited by the coupling V, which vanishes at this angle.) A similar calculation using the result of Eq. (33) is very cumbersome. In practice, we have found it preferable to solve the equations (1) numerically for a given configuration of a few hundred atoms, and then average over an ensemble of configurations. These results will be reported in a separate publication.

5. DISCUSSION AND CONCLUSIONS

In the course of our theoretical investigation of the transient behavior of an optically excited gas, we have come across a particular class of polarization waves. They are Frenkel dipolar excitons, with the difference that the degeneracies of the s and p states have been lifted by the application of a static magnetic field. First, we note that the spin-orbit coupling separates the $p_{1/2}$ states from the $p_{3/2}$ states. Then, considering the $p_{1/2}$ states, we see that the interaction between the $|s,\uparrow\rangle$ and $\left|p_{1/2},\, m_J = \frac{1}{2}\right\rangle$ states is indeed proportional to $P_2(\cos\theta)$, where θ is the angle between the interatomic vector and the applied magnetic field. In the actual experiments,[2,3] there is a spatially varying magnetic field in the magneto-optical trap, and the full analysis is much more complicated.

In the simplest case these waves are dispersionless and their frequency, given in Eqs. (6) and (9), depends on the angle θ_k between the propagation vector and the field's direction according to $1 - 3\cos^2\theta_k = -2P_2(\cos\theta_k)$. These waves have not (to our knowledge) been previously recognized, perhaps because their occurrence is confined to systems that can be described by the highly simplified Eq. (1a), which additionally is treated in the continuum approximation.

We remark that the continuum approximation becomes exact in the long wavelength limit and is expected to have a wide range of validity because the dipole-dipole interaction is long-ranged. The polarization waves also exist on a regular lattice, with a dispersion relation that can be calculated.[5,7] In a frozen gas the excitation spectrum will spread out, but localization effects are probably small, again because of the long-range nature of the dipole-dipole interaction.

Although in our model system almost all atoms are in an excited s state, they could just as well be in the ground state. What is important to the model is that this state couples effectively to a single p state. This is a good approximation for the Rydberg atoms of the system we are modeling,[2,3] but it is not a good approximation in general. Also, the size of the dipole matrix element μ is particularly large for Rydberg atoms.

Finally, we mention that the $1/r^3$ dipole-dipole interaction does not include retardation effects. However, these are expected to influence the results only by inducing some dispersion at very long wavelengths.

In this paper we consider a particular mechanism of excitation of the polarization waves, namely, we allow the resonant transfer $ss' \to pp'$ to create the initial p state that then interacts with the s states.[3] However, other mechanisms of creation of the p state would work just as well as long as a single p state is created. We have also assumed that if only a few p atoms are created, then each can be treated independently in accordance with the sparse limit.

We outline (but do not carry out in detail in this paper) the calculation of the transient behavior involving the excitation of the newly-found polarization waves in a frozen gas. While this allows us to establish contact with existing or currently planned experiments, one should perhaps think instead of more direct ways of observing the polarization waves.

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4. J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1975), p. 767. If $V(\mathbf{r})$ cuts off at small r, instead of being strictly proportional to $1/r^3$, then the radial integral in Eq. (5) will give a k dependence. A spin-spin correlation function $g(r)$ has the same effect.
5. The dispersion relation for spins on a lattice can be obtained from M. H. Cohen and F. Keffer, "Dipolar Sums in the Primitive Cubic Lattices," Phys. Rev. **99**, 1128 (1955).
6. This result can also be obtained from Eqs. (12) and (15). Note that the limit $U \to 0$, or $\mu \to 0$, corresponds to $z \to \infty$.
7. See, for instance, D. E. Logan and P. G. Wolynes, "Self-consistent Theory of Localization in Topologically Disordered Systems," Phys. Rev. B **29**, 6560 (1984); D. E. Logan and P. G. Wolynes, "Anderson Localization in Topologically Disordered Systems," Phys. Rev. B **31**, 2437 (1985); K. S. Schweizer, "Electronic Absorption of Frenkel Excitons in Topologically Disordered Systems," J. Chem. Phys. **85**, 4638 (1986).

FIGURES

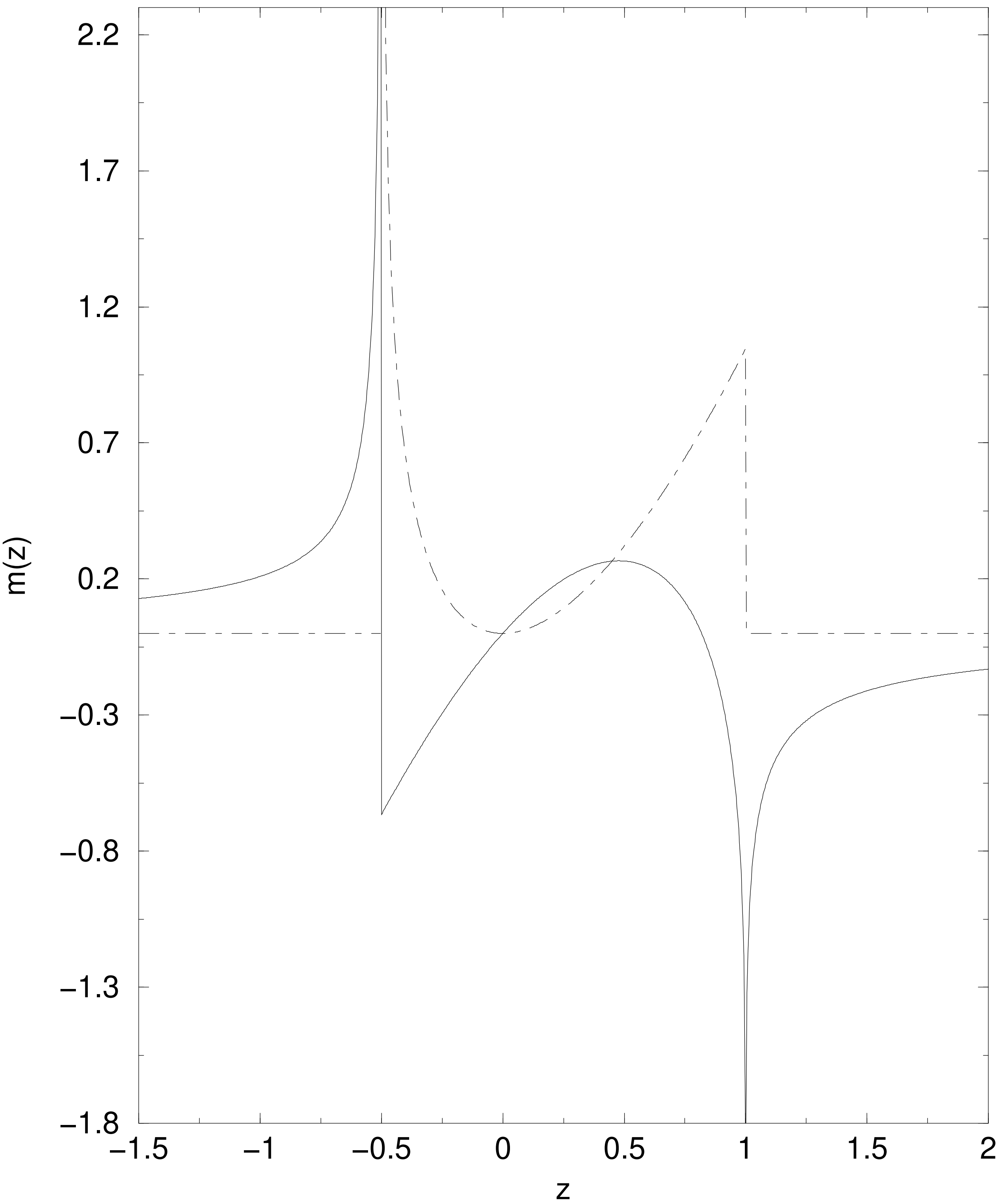
2.2
1.7
1.2
0.7
0.2
−0.3
−0.8
−1.3
−1.8
m(z)
−1.5
−1
−0.5
0
0.5
1
1.5
2
z

Fig. 1. The real part (solid line) and imaginary part (dashed line) of the function $m(z)$ for z just above the real axis.